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# Towards sustainable energy materials: broadening life cycle assessment for emerging technology development and resource-effective choices

Edis Glogic

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Edis Glogic. Towards sustainable energy materials: broadening life cycle assessment for emerging technology development and resource-effective choices. Chemical Physics [physics.chem-ph]. Université de Bordeaux; University of Waterloo (Canada), 2019. English. NNT: 2019BORD0317. tel-02869841

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THÈSE PRÉSENTÉE  
POUR OBTENIR LE GRADE DE  
**DOCTEUR DE**  
**L'UNIVERSITÉ DE BORDEAUX**

SCIENCES CHIMIQUES

CHIMIE PHYSIQUE

Par Edis GLOGIĆ

**TOWARDS SUSTAINABLE ENERGY MATERIALS:  
BROADENING LIFE CYCLE ASSESSMENT FOR  
EMERGING TECHNOLOGY DEVELOPMENT AND  
RESOURCE-EFFECTIVE CHOICES**

Sous la direction de : Guido SONNEMANN  
(co-directeur : Steven B. YOUNG)

Soutenue le 9 Décembre 2019

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**Titre :** Vers des matériaux énergétiques durables : élargissement de l'analyse du cycle de vie pour le développement de technologies émergentes et des choix économes en ressources

**Résumé :** Les matériaux énergétiques sont particulièrement intéressants du point de vue du développement durable pour faire progresser les systèmes d'énergie renouvelable, notamment les énergies de production et de stockage. Leurs utilisations appropriées ainsi que leur développement requièrent une méthode d'évaluation quantitative. L'Analyse de Cycle de Vie (ACV) est une méthode qui soutient le développement durable par l'identification de priorités environnementales ainsi que par la comparaison de différentes technologies. Cette recherche vise à soutenir le développement des matériaux énergétiques et de faire de la méthode d'analyse du cycle de vie un outil plus pertinent pour l'évaluation environnementale à travers l'extension de son usage dans deux directions émergentes : l'évaluation des technologies au début de leur développement et le soutien des choix économes en ressources dans le contexte d'une économie circulaire.

Les objectifs de recherche se focalisent sur le développement de l'information relatives aux technologies ainsi que sur la méthodologie d'identification des défis et opportunités par l'application de l'ACV sur trois études de cas de technologie énergétique à différents niveaux de maturité. Dans le premier cas d'étude, les piles alcalines, actuellement à haut niveau de maturité ( ) sont évalué grâce à l'utilisation de l'ACV combiné avec un indicateur d'économie circulaire, l'indicateur de circularité du matériau (MCI). Le but était d'explorer une opportunité de couplage des deux méthodes ainsi que les compromis entre les indicateurs pour différentes stratégies de conception et de gestion de ces batteries. Dans le deuxième cas d'étude, les électrodes à base d'hydroxyde de nickel-cobalt, à présent à bas niveau de maturité (échelle de laboratoire) sont évaluées dans l'optique d'étudier des priorités environnementales des voies de synthèse favorables. Dans le troisième cas d'étude, les chargeurs organiques photovoltaïques portables pour petits équipements électroélectroniques, actuellement à un niveau de maturité intermédiaire (échelle pilote), sont évalués pour remplacement du réseau électrique traditionnel pour le chargement de téléphones portables.

Les résultats pour la pile alcaline montrent la valeur et la portée de l'indicateur MCI pour l'évaluation de stratégies comparés aux résultats de catégorie ACV et d'indicateur. Dans ce contexte, une méthode de couplage et de présentation de l'indicateur MCI est proposée, et un besoin d'améliorer la caractérisation de la qualité des matériaux de perte pour les matériaux secondaires (recyclés) est identifié. Le cas d'étude des électrodes offre un aperçu des priorités environnementales et de leur statut par rapport aux technologies alternatives existantes, en incluant les avantages de certaines étapes de processus et voies de synthèse. Les paramètres d'opération les plus favorables en terme de densité de courant et de durée de vie espérée sont identifiés. L'analyse des chargeurs photovoltaïques montre la potentielle performance environnementale de cette technologie étant donné le contexte géographique et l'intensité d'utilisation. Les chargeurs ont prouvé leur éventuelle valeur de substitut au réseau électrique local dans trois parmi six pays en usage fréquent, et dans des catégories d'impact spécifiques. Deux contributions pour le développement et l'utilisation de l'indicateur sont complétées par

des recommandations concernant de meilleures définitions et catégorisations des indicateurs d'économie circulaires pour le cadrage des évaluations quantitatives (incluant l'ACV) et en vue de leur contribution sociale économique à l'analyse environnementale. Les cas des électrodes et des chargeurs démontrent des incertitudes relatives à l'allocation des flux de référence de l'unité fonctionnelle, qui sont gérées par la mise en place d'un scénario et une analyse de sensibilité. Étant donné les défis et la conduite des actions de réponse, les efforts devront porter sur la phase d'interprétation de l'ACV, une observation qui aurait des implications plus larges en ACV pour l'évaluation des technologies émergentes.

Les futures recherches devraient considérer comment sont utilisés les indicateurs d'économie circulaire et comment ils pourraient être étudiés avec et en complément de méthode d'allocation quantitatives comme l'ACV. Dans un contexte de modélisation de technologies émergentes avec l'ACV, il est recommandé de mettre plus d'emphasis sur la classification des ACV orientée vers le futur pour les technologies émergentes, afin de mieux cadrer et organiser les avancements méthodologiques existants et futurs dans ce domaine. Une recommandation est aussi faite au regard de l'utilisation de l'ACV attributionnel et conséquentiel pour guider le développement des technologies à travers différents niveaux de maturité.

**Mots clés :** Matériaux durables, évaluation environnementale, analyse du cycle de vie

**Title:** Towards sustainable energy materials: broadening life cycle assessment for emerging technology development and resource-effective choices

**Abstract:** Energy materials are particularly important from a sustainability perspective for advancing renewable energy systems, including energy production and storage. Their appropriate use and development require quantitative assessment methods. Life Cycle Assessment (LCA) is a method to support sustainable development that can be used to identify environmental hotspots and compare different technologies. The purpose of this research is to support development of several energy materials and make LCA a more relevant tool for sustainability assessment by extending its use in two emerging directions: assessment of technologies at the early stage of development, and by supporting more resource-effective choices for a circular economy.

The research objectives focus on informing the development of technologies and identifying methodological challenges and opportunities by applying LCA to three energy-technology case studies, each at a different technological maturity level. In the first case study, alkaline batteries, currently at a high maturity level (incumbent products), are evaluated using LCA in combination with a circular economy indicator, the Material Circularity Indicator (MCI). The aim was to investigate opportunities to combine the two methods, while considering trade-offs between indicators for different strategies for battery design and management. In the second case study, nickel-cobalt hydroxide charge storage electrodes, currently at a low maturity level (laboratory-scale), are evaluated to investigate environmental hotspots and preferred synthesis route. In the third case study, organic photovoltaic portable chargers for small electronics, currently at a medium maturity level (pilot-scale), are evaluated for replacing conventional electricity grid for charging a mobile phone.

The results of the alkaline batteries case study show the value and meaning of the MCI circular economy indicator to evaluate resource strategies as compared to LCA category and indicator results. In this context, an approach for combining and presenting the MCI indicator is proposed, and a need to improve characterization of material quality losses of secondary (recycled) material was identified. The electrodes case study offers insights on the environmental hotspots and relative status among technology alternatives, including the benefit of certain process stages and synthesis routes. The most favourable operating parameters in terms of current density and device lifetime expectations are identified. The analysis of photovoltaic chargers shows their environmental-performance potential given the geographical and use-intensity contexts. The chargers have shown as potentially valuable substitutes to local electricity grids in three of six countries given frequent use, and for specific impact categories. Case studies on electrodes and chargers demonstrate uncertainties in relation to allocation of reference flow to functional unit, which are addressed conducting scenario and break-even analysis. Given challenge and carried out responses, involve increasing efforts in the interpretation phase of LCA, an observation with potentially broader implications to the assessment of emerging technologies in LCA.

Further research should consider how circular economy indicators and could be used with and complement quantitative assessment methods such as LCA. In the context of LCA

of emerging technologies, it is recommended that more emphasis is given to further classification of future-oriented LCA studies of emerging technologies, in order to better frame and organize methodological advancements in this area. A recommendation is also made in consideration to application of attributional and consequential LCA approaches in guiding technology development at different stages of technological maturity.

**Keywords:** Sustainable materials, environmental assessment, life cycle assessment

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## ACKNOWLEDGEMENTS

I have many people and organisations to thank for enabling this research, and making it enriching and rewarding experience. I benefited of the advice, professionalism and intellectual aptitude of my mentors and colleagues, support of the family and friends who provided an important balance to my work, and funding support of organisations that enabled my research and mobility throughout these five years. Integral to creating those opportunities was a partnership with University of Waterloo in Canada where I was also fully enrolled as a Ph.D. student at Faculty of Environment, within School of Environment, Resources and Sustainability department, and Industrial Ecology group.

Primarily, I am thankful to my advisors Prof. Steven Young and Prof. Guido Sonnemann who have shown tremendous support throughout this process. Their fate, patience and encouragement helped me overcome many challenges and persevere in this long journey. They were always quick to respond with valuable feedback and advice, and were able to work with my talents and around my shortcomings. They also helped me enormously with funding and administration that were particularly challenging given joint nature of this project. In addition, they encouraged and enabled me to attend numerous academic conferences that played an important role in gaining expertise, confidence and motivation to pursue my research and understand its value. *Steve, your ability to engage with open mind and encourage building on my ideas that were sometimes very vague, were critical for pursuing this research from interdisciplinary perspective and realize its practical value. Guido, your ability to look at the research and beyond the topic itself with broader academic and institutional interests is unique, so is your leadership within our group in Bordeaux and dedication to your work. From both of you I had a lot to learn that goes well beyond my research skills and familiarity to our academic field. Thank you!*

As part of my mentoring team, I would also like to extend my gratitude to Prof. Goretty Dias, who consistently supported me throughout this process by finding time to meet and discuss my research on numerous occasions, read my work and offer excellent feedback. *Goretty, this Ph.D. journey would have been more cumbersome and less useful if I wasn't fortunate enough to have you taking part in that process. Thank you for your thoughtful advice, feedback, and moral support.*

I would also like to thank the rest of my thesis examination jury, professors Armin Reller, Laurent Servant, Jennifer Kellar, Bertrand Laratte, and Dr. Michael Tsang, who accepted to take part in my thesis defense, travel to Waterloo, and pose some challenging questions. Many thanks to Prof. Servant who joined my thesis committee at a later stage and was also very helpful with administrative aspects regarding the thesis approval and submission. I am also additionally thankful to Michael Tsang for his post-defense feedback and whose research originally served as a foundation to some of my own. Lastly, I wish to thank Prof. Stephen Quilley who was a member of my thesis committee in the early stages of my research in stages of comprehensive examination and development of my thesis research proposal.

Secondly, I would like to acknowledge two Ph.D. colleagues Steffi Weyand and Alberto Adàn Màs with who I worked closely in two of the case studies that resulted in two of the core deliverables. This had a significant impact on my success with publications, and made research very enjoyable. On that account, I am also grateful to advisors of Alberto and Steffi and authors in two papers, who encouraged this collaboration and provided feedback.

I would also like to thank my family, friends and colleagues who provided important social life-balance to my work. My brother, mother and father, grandmother, aunt and uncle, and stepmother, were with me all along this process to share all my worries and victories. I wish to also thank to all my colleagues from CyVi group in Bordeaux: Raphaël, Philippe, Baptiste, Jair, Juliene, Alice, Elkin, Javier, Dimitra, Alexander, Thibaut, Marc, and Dieuwertje. Among CyVi members, but also outside the office, I am particularly thankful to my friend and flatmate Amélie Thevenot who is one of the kindest people I have met and was both great colleague, friend and housemate. Thanks to my colleagues from IDS FunMat, particularly Uyixing, Mylene, and Mehdi, friends from Bordeaux, Nico, and Christopher. From Waterloo, I am really grateful for support to Jenny in first years of my research, my colleagues and friends Bereket, Sophie and Basak who were incredibly supporting at that particularly challenging last few months of my thesis writing. It was also great to establish deeper collegial relation with Allison and Alexander.

Of course, this research would not have been possible without several organisation who saw the importance of research on environment and sustainability, and my topic in

particular. The project was predominantly supported through University of Waterloo and University of Bordeaux within umbrella of International Doctoral School for Functional Materials (IDS-FunMat). Remainder of the funding was allocated from Prof. Young as part of the research assistantship, and secured through combination of bursaries and awards (notably, Millennium Graduate Bursary and Erasmus+), teaching appointments at University of Waterloo, and project-based work with KIC Raw Materials (Whisper project) and research collaboration with University of Poitiers arranged by Prof. Sonnemann.

Lastly, I would like to thank the people who recommended me for this Ph.D. position five years ago, professors Lilliana Rodić-Wiersma and José Potting, and Prof. Kemal Delijić for initially giving his support to my first kick-start of academic career by his initial recommendation and faith to my abilities to excel in this path.

## **DEDICATION**

Dedicated to my grandmother Julka, a retired primary school teacher who showed me that learning can be rewarding.

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## LIST OF ABBREVIATIONS

ALOP	agricultural land occupation
CALCAS	Co-ordination Action for innovation in Life Cycle Analysis for Sustainability
CE	circular economy

CED	cumulative energy demand
CFC	chlorofluorocarbon
EAF	electric arc furnace
FDP	fossil depletion
FETP	freshwater ecotoxicity
FEP	freshwater eutrophication
FU	functional unit
GO	graphene oxide
GWP	climate change
HTTP	human toxicity
IRP	ionizing radiation
ISO	International Organization for Standardization
LCA	life cycle assessment
LTP	natural land transformation
MCI	material circularity indicator
METP	marine ecotoxicity
MEP	marine eutrophication
MDP	metal depletion
NCED	nickel cobalt electrodeposited
NCED-rGO	nickel cobalt electrodeposited reduced graphene oxide
NCCP	nickel cobalt coprecipitated
ODP	ozone depletion
OPV	organic photovoltaic
PET	polyethylene terephthalate
PCBM	phenyl-C61-butyl acid methyl ester
POFP	photochemical oxidant formation
PMFP	particulate matter formation
PTFE	polytetrafluoroethylene
PV	photovoltaic
P3HT	poly(3-hexylthiophene)
rGO	reduced graphene oxide
TAP	terrestrial acidification
TETP	terrestrial ecotoxicity
TRL	technology readiness level
ULOP	urban land occupation
WDP	water depletion

# Chapter 1: Introduction

## 1.1. Research context and problem rationale

Assessment methods play an important role for moving towards sustainable products and resource use. “What cannot be measured, cannot be managed” is a phrase often used to highlight that unless there is a way to quantify the system, actions for improvement are not warranted. Within the toolbox of Industrial Ecology, the Life Cycle Assessment (LCA) method has been used to provide science-based support for environmental sustainability improvements of goods and services. In this context, LCA embodies the work of several organizations and the broader scientific community that has been central to its rapid development over past three decades. Namely, standards ISO 14040 and ISO 14044 of International Organization for Standardization (ISO), “Code of Practice” of Society of Environmental Toxicology and Chemistry, and International Reference Life Cycle Data System Handbook offer guidelines to method and procedures, support harmonization, and continuous improvements of methodology (Consoli 1993; ISO-14040 2006; ISO-14044 2006; Wolf et al. 2012). In turn, LCA has long been used and has found diverse applications in policy and industry. In policy, LCA has been used since early 2000’s (Owsianiak et al. 2018) to incorporate life cycle thinking in pollution-prevention of products (e.g., Mudgal and Benito 2008), support the introduction of new technologies, and management and taxation of solid waste (e.g., Björklund and Finnveden 2007; European Parliament and Council 2008; Meylan et al. 2015). In industry, used initially to inform packaging in the late 1980’s, LCA is now used in all aspects of product management and procurement, including the process development, marketing, monitoring of environmental performance of products and production, supply chain management, and strategic planning (Baumann and Tillman 2004; Fava et al. 2000; Guinee et al. 2010; Hauschild, Rosenbaum, and Olsen 2018).

Despite its widespread use, and significant efforts invested in harmonization and streamlining for more comprehensive and consistent assessment, LCA is criticized for lacking a true “sustainability” perspective. Mainly, it is perceived that the outlook on only environmental aspects is limited if not pursued with consideration to a broader socio-economic context. Specific critiques include the method’s ability to: address potential trade-offs between environmental and socio-economic aspects; support decisions from the perspective of sustainable production and consumption; address system dynamics and different scales of production; and forecasting of future systems (Dreyer, Hauschild, and Schierbeck 2006; Franze and Ciroth 2011; Hertwich 2005; Norris 2001; Ny et al. 2006; Reap et al. 2008; Sala, Farioli, and Zamagni 2013a).

In an effort to overcome these shortcomings, various approaches have been suggested, with discussion particularly intensifying in recent years. Efforts can be traced to the work of Andersson et al. (1998), who combine socio-ecological principles with LCA (Andersson et al. 1998). Ny et al. (2006) use "basic sustainability principles" as outlined in The Natural Step as the distance-to-target approach to account for degradation of the environment, resource use and resource sufficiency (Ny et al. 2006; Robèrt 2000). In a similar manner, Heijungs et al. (2014) use planetary boundaries in effort to provide an absolute reference to emission burdens to model how products fit within sustainable consumption (Heijungs, de Koning, and Guinée 2014). A large number of studies have also proposed combining LCA with social and economic assessment methods and indicators to complement environmental assessment (e.g., Norris 2006; Onat et al. 2016; Sonnemann, Tsang, and Schumacher 2018). More recently, Klopffer et al. (2008) combine LCA with social and cost-analysis by means of Social Life Cycle Assessment and Life Cycle Costing for an integrative Life Cycle Sustainability Assessment (Klopffer 2008). A similar, but more conceptually open approach was proposed by Guinee et al. (2010) in which a variety of methods could be used alongside LCA to address social and economic aspects of products (Guinee et al. 2010). Aforementioned efforts have extended the scope of environmental LCA even though, the adaptation of LCA in all aspects pertaining to sustainability remains an ongoing objective (Moltesen and Bjørn 2018; Sala et al. 2013a).

To transcend from its conventional focus on the environment to a method that can support broader sustainability choices, there is a need for "broadening", "deepening" of LCA, and "leaping forward" (CALCAS 2009; Guinee et al. 2010). *6<sup>th</sup> EU Framework for Co-ordination Action for innovation in Life Cycle Analysis for Sustainability* (CALCAS) differentiates two types of broadening: broadening the scope of indicators, and broadening the object of analysis. The broadening of the scope of indicators includes broadening beyond environmental assessment to include the social and economic aspects (CALCAS 2009; Guinee et al. 2010). The broadening of the object of analysis entails the use of LCA beyond product assessment, referring to company and sector (meso-level) scales of analysis, and to economy scale (macro-level) questions.

In terms of broadening LCA beyond focus on environment, a significant opportunity for LCA lies with the new sustainability paradigm of circular economy (CE), where many efforts have been directed at different levels of economy and organisations, including significant interest to bring these considerations at the product-level (Ghisellini, Cialani, and Ulgiati 2016; Haupt and Zschokke 2017). CE aims to optimize resource use, including reduced consumption of virgin raw materials and generation of waste, through closing loops in production and consumption of materials (Haas et al. 2015). Given that the concept of resources is inherently a human construct

(Dewulf et al. 2015), their preservation, localisation, and provision have socio-economic value that can be evaluated and complement the environmental analysis in LCA.

Sustainability improvement could also be supported by extending the scope of LCA use beyond conventional applications. Specifically, use of LCA to inform technology innovation and beyond ex-post (retrospective) LCA of current (incumbent) products to model future technologies is increasingly considered. Modeling of emerging technology as future systems is commonly referred to as “ex-ante LCA” (Cucurachi, van der Giesen, and Guinée 2018; Villares et al. 2017), although terms are still evolving (Buyle et al. 2019). Recent invited articles in the journal *Nature Materials* and the *Journal of American Ceramic Society* attest to that interest, for example where LCA has been identified as a key tool to assist scientific research in functional materials and energy applications (Kirchain, Gregory, and Olivetti 2017; Smith et al. 2019). However, the materials science community has not fully embraced the use of LCA due to challenges in the assessment of emerging technologies (Smith et al. 2019).

This research tries to extend the scope of LCA in two directions: broadening the scope of indicators to include indicators of a new economic paradigm of CE, and considering LCA for assessment of emerging technologies. Two new use-contexts hold a great promise for further proliferation and advancement of LCA as a development and decision-making tool, and extending the scope of the current ex-post environmental LCA. Among emerging technologies and within the context of CE, energy materials are of particular interest to materials science and industry given their role in improving renewable energy systems (Santoyo-Castelazo and Azapagic 2014).

## **1.2. Purpose and objectives**

The overall purpose of the research was to make LCA a more relevant tool for sustainability assessment of materials, by considering an integration of socio-economic indicators of circular economy to allow for resource-effective choices, and by enhancing its use for the assessment of emerging technologies.

Two research objectives were identified:

1. Evaluate methodological potential of CE-indicators to complement environmental assessment with LCA.
2. Use LCA to improve the development of emerging energy materials.

And the following specific research questions were established:

Under objective 1:

- 1-1. What are the challenges in combining LCA with circularity indicators focusing on impact-circularity trade-offs and methodological differences?
- 1-2. What methodological improvements can be suggested to address these challenges?

Under objective 2:

- 2-1. What are the environmental sustainability implications of new energy materials including opportunities/aspects for optimization across product life cycle, and when compared with conventional alternatives?
- 2-2. What are challenges and methodological approaches for improving assessment of emerging technologies?

In line with objective 2, the research considered three case study technologies: alkaline batteries, nickel-cobalt (Ni-Co) hydroxide charge storage electrodes, and organic photovoltaic solar charger, which are considered energy technologies. Objective 1 was addressed using the case study of alkaline batteries, and objective 2 was considered using the case studies of charge storage electrodes and organic photovoltaic solar chargers. The rationale for selection and characteristics of the case studies are explained in Section 1.4.1.

## **1.3. Background**

Given the purpose of this research to improve sustainability assessment using LCA, this chapter provides further background on the sustainability challenges and LCA methodology, use of LCA in context of circular economy (CE) and emerging technology assessment. The chapter starts with a description of what pertains to sustainability and what are the challenges with a particular emphasis on resource use. The subsequent section provides an overview of indicators developed to lead CE choices at the material and product levels as well as their challenges and limitations leading to the justification of the potential research gap addressed in this research. The last section of this chapter gives an overview of the assessment of emerging technology with LCA, review of the LCA case studies, serving to motivate case studies and exploration of this area.

### *1.3.1. The sustainability challenge*

Sustainability has emerged as an important direction to how resources need to be utilized with regard to economic progress, social well-being and impacts on the environment (Gibson 2005). A more sustainable future needs to prioritize intergenerational and intragenerational equity

pertaining to the availability, access, and share of resources among the human population. At the same time, resource-intensive human activity has to remain within the ability of the natural environment to absorb related pressures from emissions and disturbance so that the functioning of ecosystems and access to resources are preserved (Rockström et al. 2009). Aspects of equity and environmental protection have been adeptly captured in the definition of sustainable development set forth by Brundtland Commission in 1987 as “a development that meets the needs of the present without compromising the ability of future generations to meet their own needs” (WCED 1987).

To achieve resource provision goals, tremendous challenges persist in efforts to accommodate the quantities and pace of resource use that has been increasing steeply in both relative (per capita) and absolute terms (i.e., globally) over the past century (Krausmann, Weisz, and Eisenmenger 2016; UNEP 2010b). From around 23 billion tonnes in 1970, extraction of mineral resources more than tripled to today’s rates, now exceeding 70 billion tonnes. Steeply increasing consumption rates have potential to further increase given current unequal distribution of resources among human population. Currently, developed countries, that constitute only 20% of the world’s population, consume 80% of natural resources (Steinberger, Krausmann, and Eisenmenger 2010). Assuming the desired improvement in living conditions in developing countries over the next three decades, while pursuing similar systems of production and provision for housing, food, mobility, energy and water supply, the present rates of resource consumption would nearly triple (Schandl et al. 2016).

The primary concern over rates of resource use are their associated negative impacts on the environment. The impacts from resource extraction, production, use, trade and disposal of resources and associated commodities are the major cause of anthropogenic pollution and have implications on both local environments and global emission concentrations (Ayres and Simonis 1994; UNEP 2010a). Local implications constitute negative environmental and socio-economic impacts on local ecosystems associated with often unfair and poorly managed activities of resource extraction and mining in developing countries (IIED 2002). Global pollutant concentrations are of concern due to their effect on perturbations of the Earth’s biophysical systems, which have significant negative impact on human populations globally (Running 2012; Steffen, Rockström, and Costanza 2011; Wackernagel and Rees 1998). According to a sizable body of research in that area, we are already transgressing some of Earth’s biophysical limits to absorb certain types of pollution or a further trends of resource use would result in such transgression in a near future (Steffen et al. 2015). Imminent danger of climate change is particularly emphasised, a consequence of greenhouse gas concentrations emitted to the atmosphere mostly to meet the demand for energy that is largely sourced from fossil fuels. Raising greenhouse gas

concentrations beyond 500 parts-per-million are expected to trigger uncertain complex mechanisms and feedback loops that could result in extreme droughts, food shortages, loss of species and collapse of ecosystems (IPCC 2012).

In addition to environmental pressures, the rate of resource use is associated with potential challenges of accessibility to some minerals and metals. Presently more than 90% of all materials are derived from non-renewable resources and most demand for materials is addressed by primary production (Allwood et al. 2012; Ashby 2012; Behrens et al. 2007; Graedel et al. 2011). While the scarcity itself is still a highly debated topic (Drielsma et al. 2016), according to some estimates, shortages of metals such as zinc, copper, and indium, may be experienced within the next 50 years (Bleischwitz et al. 2009; Meadows, Randers, and Meadows 2004; Ragnarsdóttir, Koca, and Sverdrup 2012). Geological scarcity is exacerbated by the resource access and increasing dependency on a broader range of materials to fulfil the current technological needs (Graedel et al. 2013). Some of the materials important for the economy, and current and future technologies, are sourced from countries where supply is not reliable and resources concentrated at only specific geographies (Ayres and Peiró 2013), or sourcing and supply chain of raw materials associated with various social issues (Young 2018; Young, Fonseca, and Dias 2010).

Given the future perceived risks of availability, accessibility and environmental pressures, all largely associated with virgin material production, sustainable development calls for decoupling well-being and economic growth from the use of resources - dematerialization, and associated environmental impacts - impact-decoupling (UNEP 2014). The decoupling ambition stipulates an increase in resource productivity accomplished through the management and technological solutions to facilitate more efficient and effective use of resources and improved recovery to which production of energy from more renewable sources is specifically emphasized (Bringezu et al. 2004; Cleveland and Ruth 1998; Jackson 2009; UNEP 2011; Young 2001). Decoupling can occur in relative and absolute terms, depending on whether the reduction of resource use is a relative decrease in comparison with present demand, or decrease is an absolute thus able to accommodate also increase in population and affluence (Schandl et al. 2016). Under the premise of sustainable development, the rate of resources consumption has to be reduced in absolute terms and the absolute reduction should not impede material sufficiency and economic prosperity (WCED 1987).

Urgency of the climate crisis and resource productivity, resource access, and provision, may foster risky and untried techno-fix solutions that shift environmental impact burdens and have unintended negative implications (Hällström 2008). Therefore, the role of quantitative assessment methods, and LCA in particular, as the only ISO-standardized environmental quantitative assessment method suitable to measure and lead resource productive choices and

needed transformation in energy supply, cannot be overemphasised. Currently, LCA can positively influence technology’s environmental intensity through more eco-efficient solutions, and given complex environmental-profile assessment based on life-cycle and systems thinking, and interdisciplinary approach (e.g., not limited to specific impacts such as greenhouse gas emission) (Sala, Farioli, and Zamagni 2013b). However, the method’s capabilities to address social and economic aspects are not currently captured by standardized methodology (Klopffer 2008), and the scope of LCA application in various domains of science and technology development needs to increase to take full advantage of its potential.

### 1.3.2. Methodological approach of life cycle assessment

An ISO-informed LCA study conforms to a certain structure and the steps recommended in the standard (ISO-14044 2006). This largely entails that an LCA study undergoes four distinct methodological phases: Goal and scope, Life cycle inventory, Impact assessment, and Interpretation phase.

Constant iteration is carried out throughout the four phases to ensure that the goal of the study is met. The relationship between stages and possible iterations are shown in schematic in Figure 1-1, as adopted from ISO 14040 standard. Next the four stages of LCA are briefly described.

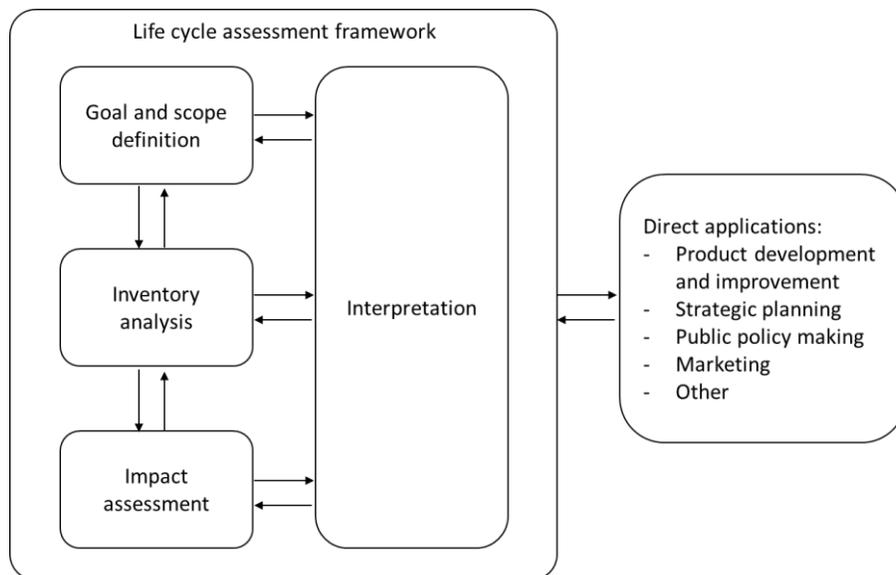


Figure 1-1. Stages of an LCA (ISO-14040 2006)

The first stage, Goal and Scope is dedicated to outlining the main purpose of a study, a unit of the analysis and the modeling properties. For the goal definition, the ISO standard mandates a definition of the study purpose, an intended audience, an application, and a declaration if the

study is to be used for comparative assertions. The scope includes: the specifications of the modelling such as the function, the functional unit and reference flow to be used as a proxy for evaluation and comparison; the boundaries of the product system and decision to include particular impacts and stages of the life cycle; specifications on the choice of impact assessment methodology and indicators; data and limitations; and a need for a critical review.

The second stage, the Inventory analysis, consists of gathering the data on relevant material and energy flow inputs and outputs of the studied product system. Foreground and background data are often differentiated. The former is collected by the analyst and the latter relied upon processes originating from the LCA databases. An outcome of the inventory analysis is a list of quantified elementary flows normalized to reference flow as determined from the functional unit.

In the third stage of LCA, Impact Assessment, inputs and outputs of materials gathered in the inventory phase are translated into potential environmental impacts. The impact assessment phase consist of several mandatory steps including: selection of impact categories and indicators that can sufficiently quantify the impacts chosen with the scope of the study, classification of elementary flows to impact categories and indicators, and characterization that entails measurement of indicator within impact category using environmental fate-exposure-effect models (i.e., cause-effect linkages estimating pollutant’s potential impact to the environmental category). Moreover, the following optional steps are foreseen: normalization of the impact assessment results to show a relative magnitude of characterized scores for each impact category, and weighting to aggregate several impact categories based on value choices to reach a single value. A description of most commonly used impact categories, is shown in Table 1-1.

The fourth and last stage, Interpretation, ensures certainty and consistency of the results are adequate and information are in a format that offers a comprehensive picture of available positions and opportunities for improvement. The interpretation phase considers choices and output from first three phases and their adequacy to fulfil the goal of a study. Sensitivity and uncertainty analyses are often applied as part of consistency, completeness and sensitivity checks, to ensure robustness of the results to potential assumptions applied to data, system boundaries, impact assessment methods, or other modeling criteria.

Table 1-1

Common impact categories in LCA (Baumann and Tillman 2004)

Category	Description
Global warming	Greenhouse gases capacity to enhance radiative forcing thereby heat the atmosphere. Several gasses with widely different capacity to absorb infrared

	radiation contribute to global warming effect including: carbon dioxide, carbon monoxide, methane, chlorofluorocarbons, nitrous oxide, and some other trace gasses. Impacts to this category are normally expresses in units of carbon dioxide equivalent (CO <sub>2</sub> eq). Given that these gases have also different life-spans in the atmosphere global warming potential is often calculated for different time horizons. Global warming potentials used in LCA are estimates developed by UN Intergovernmental Panel on Climate Change.
Ozone depletion	Impacts to stratospheric ozone layer as a result of emission of various bromated and chlorinated substances such as chlorofluorocarbons (CFC) and halons. Ozone depletion potential developed for LCA by World Meteorological Organisation, reflects on change of stratospheric column due to amount of ozone depletion emissions normalized to CFC-11 equivalents.
Toxicity	Impacts of various substances with different range and type of toxicity to human health and environment. Characterization is based on fate, exposure or intake, and effect of toxic substances while accounting for various physical and regional conditions. Toxicological data, models or empirical data. The reference substance to which toxicity of materials is estimated is 1,4-dichlorobenzene.
Photo-oxidant (smog) formation	NO <sub>x</sub> and hydrocarbon concentrations as precursor of smog that causes irritation of respiratory system in humans, and damage to vegetation.
Acidification	Concentration of air pollutants (SO <sub>2</sub> , NO <sub>x</sub> , NH <sub>3</sub> and HCl) that precipitate in form of acidic rain, fog, or snow, with damage to ecosystems and human health.
Eutrophication	Excessive concentration of phosphorus and nitrogen nutrients that can increase biological productivity and absorb oxygen having significant negative potential to aquatic and terrestrial ecosystems. These measurements reflect on biological and chemical oxygen demands. Common reference to eutrophication is PO <sub>4</sub> <sup>3-</sup> equivalents.
Land use	The use of land from the perspective of occupancy and transformation expressed in m <sup>2</sup> .
Resources	Use of resources, including renewable and non-renewable, and biotic and abiotic, involving depletion and impact.

Given the choice of background data and system boundaries delimitation, two types of LCA are commonly differentiated: the consequential and the attributional approach (Ekvall and Weidema 2004). These approaches differ in how emission burdens are shared among different co-products. The attributional approach uses partitioning to determine how burdens among co-products are distributed, whereas the consequential approach uses system expansion to include co-product systems. Accordingly, the attributional LCA is used for modeling the systems where impacts based on industry average data use is appropriate. The consequential LCA measures the impacts of the product in the context of the market and economy and various marginal effects on supply

and demand of materials motivated by raw material or product availability (Ekvall and Weidema 2004; Sandén and Karlström 2007).

The four stages of LCA described and shown in Figure 1-1, and impact categories shown in Table 1-1, were consistently carried out in all case studies in the current research (Chapters 2-4). Studies use different methods and approaches in the interpretation phase, but overall, the modeling consistency was followed. In the course of this research, both attributional and consequential methods were used.

### *1.3.3. Supporting resource-effective “circularity” choices with life cycle assessment*

One of the ways of broadening the scope of LCA and consistent with “(broadening LCA) by adding economic impacts, social impacts, or environmental impacts that are not covered by present-day LCA” (Zamagni et al. 2009, p17), is by incorporating socio-economic indicators of circular economy (CE). With the origins in the industrial ecology and industrial symbiosis concepts, CE aspires to improve resource and impact decoupling and has become widely embraced in both industry and policy spheres (Blomsma and Brennan 2017; EC 2011; EMF 2012). In comparison to conventional approach to products of take-make-use-dispose manner, the CE aspires to seize more value out of virgin resources through different product design, management and business strategies that incentives closing the loops of materials (e.g., building durable products, selling service instead of products, etc.). Moreover, cost-effective production and consumption of CE is expected to have positive implication to job creation, sufficiency and localisation of resources, thus promoting positive improvements to socio-economic and environmental aspects.

With the popularization of CE idea and identified need for framing and operationalizing this concept, the role of LCA for directing CE choices at the product level has been highlighted (CIRAIG 2015; Haupt and Zschokke 2017; Sassanelli et al. 2019; UNEP 2014). A science-based life cycle thinking approach represented through LCA is needed to ensure that resource productivity initiatives (i.e., policies and business models underpinning CE concept) reflect positively on environmental performance. Traditionally, LCA was used to inform any improvement resource strategy directed for resource minimization or recovery, either as a motivated contribution to CE or simply deemed as an environmentally-relevant strategy.

In addition to LCA that is useful to provide environmental impact evaluation of CE-motivated strategies, the need to encourage and monitor resource use conducive to CE at the product level was identified (Ghisellini et al. 2016). As a result, various indicators that encourage closing loops of materials to reduce the use of virgin resources have been proposed (see Table 1-2). Given the

focus in the current research on raw material resources, new indicators could offer new material efficiency management perspective in assessment of products alongside environmentally-relevant indicators and categories of LCA. For instance, most of circularity indicators would discourage permanent loss of materials through product end-of-life management involving incineration, and encourage disassemble or re-manufacture in design of products. Therefore, combining “circularity” indicators with established assessment methods such as LCA, Material Flow Analysis and Multi-Criteria Decision Analysis, remain an important part of their development and use (EMF 2015; Pauliuk 2018). This combination can uncover possible trade-offs between MCI and LCA indicators and categories and encourage solutions for optimal environmental performance and circularity. Integration of LCA with CE-indicators is not discussed in the work of CALCAS, which preceded development of these indicators. However, CE-indicators certainly bear resemblance to methods such as Material Input per Unit of Service (MIPS), originally discussed in CALCAS Deliverable 20 Blue paper on Life Cycle Sustainability Analysis (Zamagni et al. 2009), and with significant traction of CE in the past decade this is increasingly of interest.

Current indicator and index approaches aimed at product-level circularity assessment are shown in Table 1-2.

Table 1-2.

Overview of product-level circularity indicators

<b>Indicator and source study</b>	<b>Description</b>
Material Reuse Indicator (Park and Chertow 2014)	Provides characterization of end-of-life products as resource-like or waste-like based on context and technological ability of materials to be reused. Demonstrated at a case of coal combustion byproduct.
Circular Economy Index (Di Maio and Rem 2015)	Represents the ratio of the monetary value between secondary material (from recycling facility) and intrinsic value of material entering recycling facility.
Material Circularity Indicator (EMF 2015)	Measures the extent of material circularity versus linearity by accounting for different material characteristics and fractions along life cycle stages of a product. MCI is determined by measuring the quantity of virgin (or secondary) material used in the product manufacture, the product use efficiency, and how much material is recovered at product’s end-of-life. Demonstrated for a washing machine and a power drill.

Longevity Indicator (Franklin-Johnson, Figge, and Canning 2016)	Shows a length of time material is retained in Technosphere taking into account: initial lifetime, earned refurbished lifetime and earned recycled lifetime. Demonstrated for precious metals in mobile phone handsets.
Displacement Rate (Zink, Geyer, and Startz 2016)	Calculates how much of secondary material (reused or recycled) would displace primary material. Displacement rate is calculated using partial equilibrium modeling accounting for five price response parameters of supply and demand for materials. Demonstrated in example of aluminium recycling.
Value-based Resource Efficiency (Di Maio et al. 2017)	Represents the ratio between input and output value of "stressed" resources - those that are geologically or market scarce, or extracting them creates externalities.
Circular Economy Performance Indicator (Huysman et al. 2017)	Indicator is based on technical quality, calculated by dividing actual benefit with ideal benefit according to quality of waste stream evaluated using Cumulative Exergy Extraction from the Natural Environment (CEENE) method. Demonstrated at a case of post-industrial plastic waste from extrusion process.
Product-Level Circularity Metric (Linder, Sarasini, and Loon 2017)	Represents the ratio between economic value of recirculated parts and economic value of all product parts. Demonstrated for a plastic toy and a starter engine.
Ease of disassembly (Vanegas et al. 2018)	Measures time for disassembly by sequence of actions specific to product or product components. The premise is that fast disassembly will increase the economic viability of product life extension techniques through repair and reuse, or improve recycling yield. Indicator demonstrated in a case of LCD monitor.

As shown in the Table 1-2, circularity indicators entail very diverse perception of how circularity of products is accomplished. Most of the CE indicator approaches focus on the end-of-life stage of products and provide a characterization of recycling or recovery practices. Other approaches that consider all stages in the product life cycle and account for market forces, such as value or context of reuse in the industry, are suggested for Displacement Rate and Material Reuse Indicator, time duration as a proxy of product durability (i.e., Longevity indicator), product disassembly (i.e., Ease of disassembly), and material quality based on ratio between original materials used in the product and the value of recovered material (i.e., Circular Economy Index, Product-Level Circularity Metric and Value-based Resource Efficiency). Several studies discuss the differences between proposed indicators based on their mechanisms, scientific validity, or anticipated actions for the transition to CE (Elia, Gnoni, and Tornese 2017; Linder et al. 2017;

Saidani et al. 2017), or according to their use for CE strategies and measurement scope (Moraga et al. 2019).

Among characterization studies, and studies proposing indicators and demonstrating indicator uses, little or no attention has been dedicated to characterization of the indicators in their practical use and conceptual value with some of the existing assessment methods, even though these aspects are considered important in their use and development (EMF 2015; Pauliuk 2018). Particularly, vastly diverse conceptualizations of the indicators also impact the abilities and opportunities to be used with these methods. From a practical perspective, circularity indicators mandate different type of data and are useful in the context of different type of strategies. From a conceptual point of view, depending on the methods that they complement, circularity indicators - given the variety of answers they support - contribute different value to existing methods. Despite these observations, thus far only three studies investigated combining results or aspects of trade-offs between circularity and environmental categories and indicators, or their joint interpretation with LCA (Lonca et al. 2018; Niero and Kalbar 2019; Walker et al. 2018).

#### *1.3.4. Assessment of prospective emerging technology development with life cycle assessment*

This section explores the use of LCA for assessment of potential future systems, particularly emerging technologies, and tries to explore research opportunities in this area by looking at current LCA case studies assessing emerging technologies and literature that addresses challenges in modeling of emerging technologies.

Use of LCA for emerging technology development attempts to inform design improvements before technology is locked in a product form and limitations to design adaptations have set in (Collingridge 1982). Given that around 80% of environmental impacts of the product are determined in the design phase (Tischner et al. 2000), such improvements are believed to be significant (Villares et al. 2017; Wender et al. 2014). For new technologies, the degree of design freedom is believed to decrease throughout technology development, whereas knowledge of technology performance and inputs and outputs of energy and materials through technology optimization and scale-up increases (Figure 1-2). Inverted and proportional to the knowledge is an uncertainty dealt within LCA models that is high at the beginning and reduces with stages of development.

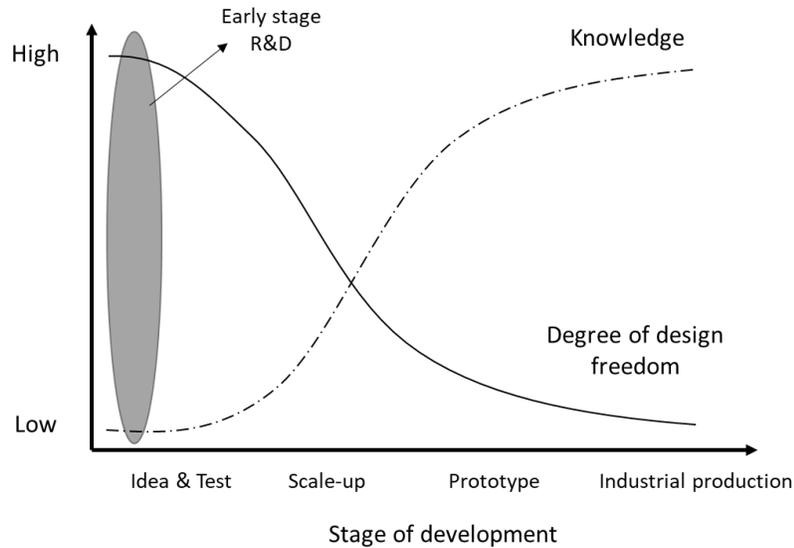


Figure 1-2. Scheme of relationship between design freedom and technology diffusion, knowledge of technology and associated uncertainty. Adapted from (Zschieschang, Pfeifer, and Schebek 2012).

Given these scale-up mechanisms, the assessment of emerging technologies in LCA need to overcome numerous challenges that increase uncertainty, challenge reliability of the results from the analysis, and affect how the LCA method need to be applied and results interpreted. Commonly reported challenges are:

- i.* The access and forecasting of data on material and energy use,
- ii.* The capability of LCA impact categories and impact assessment methods to deal with new types of materials and associated pollutants (e.g., characterization factors to estimate the fate, transport and effect of nanomaterials (Gilbertson et al. 2015; Upadhyayula et al. 2012)),
- iii.* Definition of function, functional unit and reference flow given the new or uncertain functionality (i.e., no reference (benchmark) for the technology, or the knowledge of how the new technology will be deployed) (Pourzahedi et al. 2018),
- iv.* The uncertainty associated with deployment of the new technology on the market (Hetherington et al. 2014), and
- v.* Challenges to carrying out contribution and comparative analysis given process materials and energy scaling-up potential (Gilbertson et al. 2015).

Scaling-up challenges in LCA refer to the uncertainty associated with the potential of reagents and energy quantities to change (reduce) from laboratory-scale synthesis (of early technology) to mass production (inherent to mature (incumbent) technology) and should not be confused with

different scales (objects of analysis) at which LCA can be applied (i.e., product, organization, sector, and economy).

To address some of the noted challenges, various approaches are proposed to adapt and complement the method to minimize uncertainties and maximize the value of the analysis. In that regard, LCA is either conducted with more extensive sensitivity and scenario analysis, or combined with other methods to resolve uncertainties and aspects not viably captured by conventional LCA. The sensitivity and scenario-based approaches include: predictive scenarios and scenario ranges for modeling of foreground and background systems (Arvidsson et al. 2018); scenarios generated from integrated assessment models (Mendoza Beltran et al. 2018); adoption of innovation to construct scenarios with anticipated market mechanisms (Sharp and Miller 2016); linear regression and cross validation to project efficiencies and fill in data gaps (Meng et al. 2019); global sensitivity analysis to address uncertainty of inventory inputs (Lacirignola et al. 2017); pre-screening for the most relevant processes to focus on the analysis (Ravikumar et al. 2018); and base scenarios on using scale-up projections (Gavankar, Anderson, and Keller 2015). Examples of methods coupling LCA with other methods to improve on uncertainties include design of experiments technique (Rivera and Sutherland 2015), and material criteria decision analysis and risk assessment to support assessment and associated uncertainties related to impact of nanomaterials (Scott et al. 2016; Sonnemann et al. 2018; Tsang et al. 2014). Discussion is also set around what an LCA of emerging technologies should entail (i.e., what type of questions can it answer), and how a level of technology maturity should be reported (Arvidsson et al. 2018; Gavankar, Suh, and Keller 2015).

Literature was reviewed to identify challenges involved in LCA dealing with emerging technologies, in order to get a sense of current progress in the area. The review was carried out in Scopus using the key words: “emerging”, “technology” and “life cycle assessment”, focusing on applications involving synthesis of materials and components while excluding emerging technologies for waste treatment, studies on biological systems (e.g. biofuels), and conference papers. From 254 initial items, 13 studies were identified as relevant. The review was realized by tracking reported challenges and methodological choices (i.e., system boundaries, impact categories, etc.) reported in case studies, and in particular the following:

- a) Technology, authors and year
- b) Scope of the study
- c) Challenges to Functional Unit (FU) reported or implied
- d) Technology Readiness (Maturity) Level (TRL) reported or implied
- e) TRL of benchmark technology
- f) Type of LCA reported

- g) If use of nanomaterials is reported
- h) If scale-up challenges are reported
- i) Nature of sensitivity or scenarios analysis
- j) Impact categories and methods used

Three criteria require further explanation for their use: consideration of TRL, the type of LCA reported, and observation to nano-materials in reviewed studies. TRL is a feature that defines the level of a technology's development and has implications for scaling-up challenges (Gavankar, Suh, et al. 2015). Use of nanomaterials is considered as indication of how challenges to (and lack of) characterization factors for nanomaterials fate and transport are prevalent in these studies, and given that the new generation energy technologies increasingly rely on use of nano-materials. The type of LCA refers to differentiation between "anticipatory", "prospective", and "ex-ante" LCA. The identification between three terms in reviewed studies suggests the level of adoption of new terms among reviewed case studies that may further hint on what is perceived as belonging under given classification and what is its overall usefulness. Distinctions between these terms is still fuzzy in the literature and with certainly some overlap. The most established term is "prospective" LCA, which traditionally refers to general distinction between whether present or future systems are modeled, and as distinct from conventional retrospective LCA. Moreover, a term "prospective" was also used to refer to consequential approach in LCA in distinction to attributional as "retrospective", although it was eventually established that both attributional and consequential approaches can be both prospective and retrospective (Curran, Mann, and Norris 2005). Arvidsson et al (2018) suggest a definition of "prospective" LCA that focuses specifically on emerging technology. According to the authors, study is "prospective" when an emerging technology is modeled as if it was a future, more mature technology. This is considered with the use of predictive scenarios (Arvidsson et al. 2018). Anticipatory LCA is a non-predictive tool inclusive of uncertainty that explores a spectrum of possible future scenarios to determine those that may be most environmentally promising for future research. Uncertainty is increased through inclusion of multiple social perspectives through stakeholder engagement, and prospective modeling tools. As such, anticipatory LCA tries to embody core principles of responsible research and innovation. Anticipatory and prospective LCA appear to be similar on the basis of questions being addressed, object of analysis (both focus on emerging technology), the scope, complementary methods used in modeling, and allocation procedure (Guinée et al. 2018), although their differences are highlighted in the degree and type of communication to carry out two modes (Pourzahedi et al. 2018). Inclusion of stakeholder values and social perspectives are not a prerequisite for the prospective LCA. Cucurachi et al. (2018) define conditions for ex-ante LCA, in that it should 1) include scale-up assumptions of future technology performance, and 2) carry out comparison with incumbent technologies. Hence the ex-ante LCA

can fit under the broader umbrella terms of “prospective”, “consequential”, “dynamic”, “anticipatory”, and “mixed” LCA (LCAs including mixed features of previous modes) (Cucurachi et al. 2018). The three modes do not capture an exhaustive list of future-oriented LCA modes associated with an emerging technology. Other modes used to model potential future products include: “dynamic”, “back-casting”, “consequential”, “scenario-based”, and newly proposed, “explorative” (Cucurachi et al. 2018; Fukushima and Hirao 2002; Guinée et al. 2018). However, these modes are not accounted for here as they also present approaches that concern existing commercial (incumbent) technologies.

A number of LCA studies that address emerging technology were identified. These studies are described in Table 1-3 and the following observations are made:

- All studies were carried out using cradle-to-gate boundaries (i.e., only production stage in technology life cycle is modeled)
- More than half of the studies were carried out in the last two years.
- Classification of LCA studies into “prospective”, “anticipatory” and “ex-ante” seems to be only taking a hold in recent years with the majority of studies not employing any of the proposed terms.
- Two studies reported uncertainty related to technology deployment, as an aspect of functional unit and reference flow definition.
- Five studies report the use of nano-materials, although potential uncertainties related to availability of characterization factors of the impact assessment methods are not reported and explored as part of sensitivity or uncertainty analysis.
- Aspects of the emerging nature of a technology are discussed in roughly half of the studies. Nevertheless, not all cases carry out scenario, sensitivity or uncertainty analysis with respect to the variables around the emerging technology; in some instances, these additional analyses were carried out but not discussed as part of the data used to describe the emerging technology. In such cases, uncertainty related to efficiencies are investigated more frequently in comparison to uncertainties related to inventory data.
- There does not seem to be a trend of using specific environmental impact categories or indicators/methods.

Together this review shows inconsistencies in how LCA is used for assessment of emerging technologies. Observations from the reviewed case studies include: the time-period of case studies, inconsistencies with LCA-mode terminology or absence of it, challenges to definition of functional unit and reference flow, reporting on characterization factors associated with novel emissions, and pursued uncertainty and sensitivity analysis that incorporate characteristic of emerging technologies. Particularly, improvements are needed to further clarify the features and

challenges of modeling emerging technologies and build on them to provide recommendations and delineate emerging technology LCAs. These observations are consistent with a recent review of emerging technology LCA case studies, which also highlights the lack of attention given to technologies at very early stage of development among emerging technologies (Buyle et al. 2019).

Table 1-3

Review of LCA studies on emerging technologies from year 2000 up to December 2018

Technology & authors	Study scope	Reported or implied challenges to FU	Reported or implied TRL	TRL of benchmark	Reported type of LCA	Reported use of nano-materials	Reported scale-up challenges	Sensitivity or scenarios addressing emerging nature	Impact categories/method used
Carbon nanofibers (Khanna, Bakshi, and Lee 2008)	Cradle-to-gate	Yes	Reported lab-scale	Conventional	None	Yes	Yes, materials data	Sensitivity to system boundary	GWP, HTTP, ODP, POFP, freshwater ETP, terrestrial ETP, AP, EP, Endpoint: HH, ecosystems, resources
Quantum dot photovoltaic module (Şengül and Theis 2011)	Cradle-to-gate	No	Implied - pilot	Conventional	None	Yes	No	No	EPBT, CED, GWP, HM
Perchlorate drinking water treatment technology (Choe et al. 2013)	Cradle-to-gate	No	Not clear	Conventional	None	No	No	No	TRACI 2.1.
Carbon nanotube-enabled chemical gas sensor (Gilbertson et al. 2014)	Cradle-to-gate	No	Reported lab-scale	None	None	Yes	No	No	TRACI 2.0.
Electronics display (Amasawa et al. 2016)	Cradle-to-gate	No	Implied - pilot	Similar TRL	None	No	Yes, materials data	No	CED, GWP
Copper recovery by bioleaching (Villares et al. 2016)	Cradle-to-gate	No	Reported lab-scale	Conventional	None	No	Yes, materials data	Scenarios to materials scaling-up	TRACI 2.0.
Chlori-alkali (Garcia-Herrero et al. 2017)	Cradle-to-gate	Yes	Implied - emerging conventional	Similar TRL	None	No	No	Sensitivity to efficiency	AP, HH-carc, ODP, POFP, aquatic AP, aquatic oxygen demand, ETP, EP, land impacts from waste

Technology & authors	Study scope	Reported or implied challenges to FU	Reported or implied TRL	TRL of benchmark	Reported type of LCA	Reported use of nano-materials	Reported scale-up challenges	Sensitivity or scenarios addressing emerging nature	Impact categories/method used
Ultra-high pressure homogenisation for milk treatment (Valsasina et al. 2017)	Cradle-to-gate	No	Reported, pilot scale	Conventional	Prospective	No	Yes, materials data	Sensitivity to efficiency	ReCiPe Midpoint E
Epitaxial graphene (Arvidsson and Molander 2017)	Cradle-to-gate	Yes	Yes - lab scale	None	Prospective	No	Yes, materials data	Scenarios to materials scaling-up	CED, GWP, terrestrial AP, ETP
Single walled carbon nanotube cell and tandem photovoltaic cell (Celik et al. 2017)	Cradle-to-gate	No	Implied lab-pilot	Similar TRL	Ex-ante	Yes	Yes, efficiency	Sensitivity to lifetime	TRACI, EPBT
Hydrogen production from natural gas (Salkuyeh, Saville, and MacLean 2017)	Cradle-to-gate	No	Implied - pilot	Conventional	None	No	No	Sensitivity to efficiency	Direct CO2 emissions
Paving blocks from bauxite residue (Joyce et al. 2018)	Cradle-to-gate	No	Yes - lab-scale	None	Anticipatory	No	Yes, materials data	No	GWP, AC, PMFP, POFP, ODP, HT-cancer, HTPP-non-carc, HTPP-non-carc, IRP, freshwater EP, marine ETP, freshwater ETP, ADP
Nanocrystal solvent (Tsang et al. 2018)	Cradle-to-gate	Yes	Reported lab-scale	Conventional	Anticipatory	Yes	No	No	ReCiPe Midpoint (H)
Thin film copper indium gallium (di)selenide photovoltaic modules (Amarakoon et al. 2018)	Cradle-to-gate	No	Reported pilot-scale	None	None	No	Yes, efficiency	No	TRACI 2.1.

Abbreviation of categories and indicators: land occupation (LOP), climate change (GWP), fossil depletion (FDP), ecotoxicity (ETP), eutrophication (EP), human toxicity (HTTP), ionizing radiation (IRP), metal depletion (MDP), natural land transformation (LTP), ozone depletion (ODP), particulate matter formation (PMFP), photochemical oxidant formation (smog) (POFP), acidification (AP), ecotoxicity (ETP), water depletion (WDP); energy payback time (EPBT), cumulative energy demand (CED), heavy metals (HM), abiotic resource depletion (ADP), human health (HH). ReCiPe Midpoint: agricultural LOP, GWP, FDP, freshwater ETP, freshwater EP, HTPP, IRP, marine ETP, marine EP, MDP, LTP, ODP, PMFP, POFP, terrestrial AP, terrestrial ETP, urban LOP, WDP. TRACI: AP, ET, HH air, HH carc., HH non-carc, EP, ODP, POFP, ADP-fossil fuels.

## 1.4. Research design

This section introduces the three case studies that were analyzed. The section provides reasoning for the case study selection in view of the objectives and research questions of this research (outlined in section 1.2.). Selected cases represent examples of less conventional application of LCA bringing value to the method development as it provides a proof of concept and reveals potential challenges (Baumann and Tillman 2004). This has been noted for both the assessment of emerging technologies with LCA (Hetherington et al. 2014), and the development of circularity indicators (Elia et al. 2017).

The cases were chosen in the general domain of energy materials, and where industrial and materials science research interests exist. Furthermore, the selection was driven on basis of several criteria including: technology readiness level (TRL), life cycle phase, access to informed actors, and applied method in LCA (i.e., differentiation between “attributional” and “consequential approaches, described in Section 1.3.2). Therefore, the intention was to have cases representative of different life cycle stages, different TRL, and to test use of both LCA methods. Proposed criteria were meant to enable more diversified reference and improve the potential for generalization. For example, in reference to different life cycle stages, having both production-phase and use-phase addressed through two case studies allowed to identify broader range of challenges in assessment of emerging technologies. Three case studies: alkaline batteries, nickel-cobalt (Ni-Co) hydroxide charge storage electrodes and organic photovoltaic (OPV) solar chargers correspond to a cross-section of these criteria as shown in Table 1-4.

Table 1-4

Aspects considered for selection of case studies in this research

		Alkaline batteries	OPV chargers	Charge storage electrodes
Focus life-cycle stage	<i>Production</i>			*
	<i>Use</i>		*	
	<i>End-of-life</i>	*		
LCA method	<i>Attributional</i>	*		*
	<i>Consequential</i>		*	
Technological maturity	<i>Lab-scale</i>			*
	<i>Pilot-scale</i>		*	
	<i>Industrial-scale</i>	*		
Informed actors	<i>Policy</i>	*	*	
	<i>Industry</i>	*	*	*
	<i>Consumer</i>		*	

In line with the purpose and the objectives of the research, the cases are in the domain of energy materials which are important from a sustainability perspective for advancing renewable energy systems (Santoyo-Castelazo and Azapagic 2014), and in the scope of this research project, represent emerging technologies and incorporate challenges to resource use (relevant to closing loops in CE). Energy storage and production technologies based on renewables sources of energy that can compete with conventional energy sources are intensively studied and developed. The new technologies are only viable if they offer greener and overall better substitutes to current energy supply systems. As we move from carbon-intensive mineral fuels to more extraction-intensive mineral resource, it is important that emissions burdens are not shifted from lower greenhouse gas emissions to new type of emissions related to toxicity or heavy metals. Furthermore, a shift to renewables strain supply of resources such as lithium (in batteries), neodymium (in magnets for wind turbines), cobalt (in lithium ion batteries), silica (photovoltaics), etc. notwithstanding the challenges from the perspective of disposal and closing loops of end-of-life materials.

OPV and charge electrodes were selected as cases for evaluating use of LCA for emerging technologies, while assessment of circularity uses the alkaline batteries case, as it is a more established technology where data of all life cycle stages are available. The position of the selected two case studies in use-contexts of LCA use investigated in this study is depicted in Figure 1-3. The case of alkaline batteries contributes to adding CE indicators to LCA impact category and indicator results, moving from environment-oriented assessment in LCA to also address other resource impacts. The cases of electrodes and OPV chargers pursue LCA assessment at different levels of technological readiness. Electrodes are positioned earlier in comparison to OPV charger given their lower technological maturity level in comparison to pilot-level OPV technology.

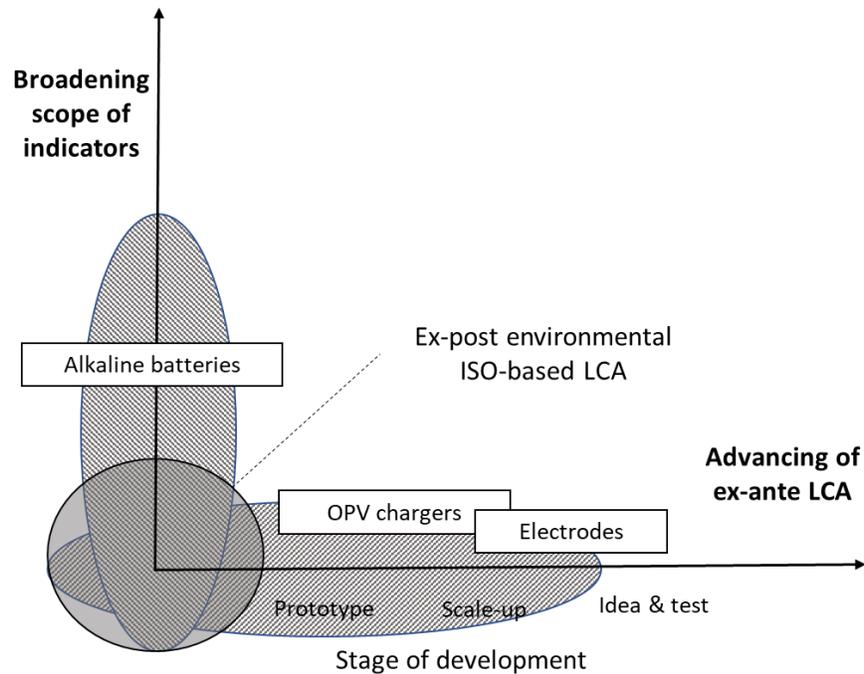


Figure 1-3. Conceptual framework of two use-context of LCA use in this study and how case studies are situated on broadening of scope of indicators (vertical scale) and looking at emerging technologies of different readiness level of ex-ante LCA

The assessment of alkaline batteries entails combining LCA methods with the so-called “Material Circularity Indicator” (MCI), which was selected among CE-indicators to assess CE performance of batteries. While other choices were available (see Table 1-2.), this particular indicator was selected given its construct and popularity. The MCI integrates a whole-life cycle approach, including multiple criteria that allows range of circularity strategies to be tested for alkaline batteries, and is frequently considered in research in the domain of CE (Azevedo, Godina, and Matias 2017; Elia et al. 2017; Saidani et al. 2017). Furthermore, the MCI can be easily calculated using data obtained in the LCA inventory. The founders of the indicator noted that MCI could be one of the parameters considered “as an output from an LCA or eco-design approach alongside those already typically used” (EMF 2015, p11).

Consistent with the broadening approach adopted in this study, it is necessary to consider the socio-economic character of the MCI indicator and its capacity to complement environmental analysis in LCA. In that regard, it has been argued that although the relationship of MCI to the three sustainability pillars is not explicit (Saidani et al. 2017), the MCI indirectly appeals to sustainability through its life cycle thinking approach and ability to measure strategies for preservation of product, components, materials, and embodied energy (Moraga et al. 2019). These strategies aim to improve resource productivity, prompt redesign of products to give more

attention to their end-of-life recovery and waste management, while also creating market demand for secondary materials. This preservation of material resources, is geared to human welfare and have therefore a strong socio-economic character (Dewulf et al. 2015). In addition, the iterations to conventional production and consumption practices implied through these strategies, and CE more broadly, are expected to have indirect implications to employment, and improve the access of resources for industry and economy (e.g., resource localization and sufficiency). Details on the methodology and calculation of MCI are described in the methodology section of the case study (see Table 2.1).

This chapter has situated the research by providing background, current literature and research gaps, corresponding research objectives and questions, and the research approach to selecting the three case studies. Two general research gaps were identified to motivate advancements in two use-contexts of LCA: (i) the lack of experience and direction on combining CE indicators with LCA, and (ii) a general weakness in guidance available on applying LCA to emerging technologies. The subsequent chapters 2-4, each present a different case study with its own introduction, methods, results, discussion and conclusion sections. Chapter 2 reports on the case study on alkaline batteries, followed by the case study on charge-storage electrodes in chapter 3, and the case study on photovoltaic chargers in chapter 4. Chapter 5, reflects on how the overall objectives and research questions were answered given the results from the case studies. Chapter 5 also gives an overview of the main contributions of the research, considers implications, discusses its limitations, and offers recommendations for future research.

## **Chapter 2: Confronting challenges in combining Material Circularity Indicator with Life Cycle Assessment: Learnings from a case of alkaline batteries <sup>1</sup>**

### **Abstract**

Product-level assessment indicators and methods are needed to incorporate circular economy ideas of resource minimization and cycling in the product manufacture, use, and end-of-life. Strategies applied to improve product circularity are only plausible if they also contribute positively to the product's environmental performance or trade-offs to the environment are acceptable. In the current work, we investigate the trade-offs between prominent circularity indicator Material Circularity Indicator (MCI) and categories and indicators of Life Cycle Assessment (LCA) in order to scope their application and improve their combined use. The methods are used to evaluate several scenarios for design and management of single-use alkaline batteries involving strategies of recycling, use of recycled content, end-of-life collection, and improved use efficiency. In addition, the trade-offs are observed under changing boundary assumptions in order to determine how the lack of quality characterization of secondary material in assessment of MCI affects the robustness of the dual analysis and applicability of MCI to specific strategies. Results suggest that trade-offs between MCI and LCA categories and indicators could be significant given the choice of recycling route and recycled content in battery manufacture (i.e., using 10% of recycled content increases MCI (9%) but also impacts (up to 6.85 %). The robustness of the results is notably affected under truncation of system boundaries to exclude byproducts of recycling in which case MCI is more significantly affected than categories and indicators in LCA highlighting the need for the characterization of material quality losses in the evaluation of MCI. We offer a new approach to visualize and identify trade-offs between indicators and compare circularity strategies.

### **2.1. Introduction**

More sustainable use of resources requires development and adequate implementation of various management strategies to be applied at different stages of the product life cycle, i.e., from

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<sup>1</sup> A version of this chapter was submitted to the journal *Resources, Conservation & Recycling* as: Edis Glogic, Guido Sonnemann, Steven B. Young. Confronting challenges in coupling Material Circularity Indicator with Life Cycle Assessment: learnings from a case of alkaline batteries.

sourcing of raw materials, product manufacture, use, and end-of-life. Strategies can be pursued to improve product's performance on desired parameters such as costs, reduction of impacts to the environment, and social aspects of resource use. Recently, they are pursued to improve resource productivity consistent with the closing material cycles in circular economy. The implementation of circularity strategies presents an opportunity to conserve the resources, but also a risk if they undermine other sustainability aspects. Primarily, decisions aimed to improve circularity should incorporate strong environmental value system and ensure closing loops of resources do not result in significant negative trade-offs to the environment (Ghisellini et al. 2016; Haupt and Zschokke 2017; Kalmykova, Rosado, and Patricio 2015).

A notable approach to quantify the influence of different strategies, aimed at improving the circularity of products, is made using Material Circularity Indicator (MCI). The indicator is developed by Ellen MacArthur Foundation and Granta Design Ltd. and integrated in to the software package MI:Product Intelligence (EMF 2015). As a multi-criteria whole life-cycle approach, the indicator is particularly popular in the industry and is among the most prominent choices for measuring circularity of the products (Elia et al. 2017). MCI measures “the extent at which linear flows of resources, used in the product have been minimised and restorative flows maximised, and how long and intensively the product is used compared to a similar industry-average product” (EMF 2015). The MCI quantifies material flow fractions and product use characteristics to reach a single value. Three main parameters are quantified to determine the MCI value: quantity of primary material used to manufacture a product, quantity of material that ends up as waste, and how long or intensively product is used (a product's “utility”). These parameters are responsive to a vast range of resource productivity and minimization strategies that can be implemented to increase circularity.

To identify the trade-offs between resource circularity and environmental impacts, MCI analysis could be complemented with life cycle assessment (LCA), an established methodology for the assessment of impacts to human health, ecosystems, and resources (ISO-14040 2006). Combining of methods would ensure that selected circularity strategies improve product circularity without significant trade-offs to the environment. Evaluation of MCI as part of the LCA framework has also been suggested by the indicator founders given the easiness of evaluating MCI as part of the LCA study, with such attempts observed for other resource-based and circularity indicators (Adibi et al. 2017; Sonnemann et al. 2015). However, the combining is challenging due to different methodologies to calculate indicator values, different valuation scales (i.e., MCI values are absolute and unitless) and means to resolve potential trade-offs. Three previous studies attempted to advance on these aspects. Lonca et al. (2018) compared environmental impact categories and indicators with MCI to assess circularity strategies for life-extension and

manufacture of truck tires. Environmental trade-offs were noted for the life-extension strategies, while the use of secondary material for tire manufacture was shown beneficial for both impact and circularity indicators. Combining and comparing indicators were facilitated by inverting MCI value to calculate “material linearity” (Lonca et al. 2018). A study by Neiro and Kalbar (2019) combined MCI with LCA categories to compare hypothetical alternatives for beer packaging. The authors proposed resolving trade-offs through multi-criteria decision analysis to enable coupling of the methods in which case circularity and environmental impact categories are weighted to reach a single score. The authors use TOPSIS method to identify the numerical distance from Positive Ideal Solution and Negative Ideal Solution, thus enable MCI (benefit-type) and LCA results (cost-type) to be compared (Niero and Kalbar 2019). Lastly, Walker et al. (2018) compared several circularity indicators (including MCI) with carbon footprinting indicator. Improvement of both MCI and greenhouse gas reductions are observed with improvement scenarios for a tidal turbine considering scenarios incorporating additional energy recovery from end-of-life product, refurbishment, and extended product lifetime. Degree of correlation between MCI and LCA categories and indicators was observed, although the authors note that MCI was unable to recognise true benefits of some scenarios that had a more significant impact on greenhouse gas reductions, in comparison to more moderate improvements of circularity (Walker et al. 2018). Although trade-offs were observed in this study, the challenges of methods combining and joint interpretation were not discussed. Combining of other circularity indicators with LCA was observed in a study by Adibi et al. (2017) in which case LCA indicator of abiotic depletion potential was coupled and compared with Global Resource Indicator incorporating recyclability and criticality to support the assessment of resources (Adibi et al. 2017). However, given the construct and nature of the indicator, the coupling is more straightforward than for multi-criteria and material efficiency-based MCI.

As a continuation of these efforts, the objective of this study is to investigate the trade-offs between environmental impact categories and indicators and MCI in order to improve their joint use and discuss the context of MCI use and development. We look more thoroughly at how the robustness of results is affected by the lack of characterisation of material quality losses (byproducts of recycling) in the calculation of MCI. The circularity and environmental analysis are conducted for several strategies for management of single-use zinc-manganese alkaline batteries in the Canadian province Ontario. Circularity strategies including recycling, the use of recycled content in battery manufacture, batteries’ end-of-life collection, and adaptations of design for improved battery performance, converge into several scenarios for battery management based on the current and prospective best industry practices and policy targets.

## 2.2. Materials and methods

### *2.2.1. Case study: challenges to closing loops of alkaline batteries*

Alkaline batteries are selected as a product with an obvious appeal from analysis of circular economy and impacts on the environment. Their intensive and widespread use, single-use life cycle, and small size all impose challenges to closing material loops. Disposal of spent alkaline batteries to landfill is discouraged or restricted due to presence of potentially toxic materials (Eisler 1993, 1998), preference to the recycling (Fisher et al. 2006), and batteries indirect role in recycling rates of other battery streams which are undermined if the disposal to landfill is permitted (Xará, Almeida, and Costa 2015).

In Ontario, a province of 14 million, these challenges pertain to around six million batteries and 5 000 metric tons of battery waste generated each year (Stewardship Ontario 2016). Collection and recycling of batteries at their end-of-life is the responsibility of battery manufacturers who fund and coordinate with recyclers to achieve particular recycling and collection rates. By provincial legislation, recycling rates of collected batteries are set at minimum 50%, while the collection of batteries is currently close to 50% and aspired to further increase (Stewardship Ontario 2016). A sizeable portion of secondary batteries in Ontario is processed by two recyclers: Inmetco and Raw Material Company, who are relevant for our investigation as their production data and the case was made available for this analysis in light of potential changes in stewardship share of the market and new circular economy policies. Recycling of batteries by Inmetco (route #1) employs mechanical and pyrometallurgical treatment, and recycling by Raw Material Company (route #2) employs mechanical and hydrometallurgical treatment. Companies produce intermediate materials that are further processed by other industries.

Given the number of battery-specific management challenges from the perspective of circular economy and impact on the environment, which are also sufficiently complex and diverse in view of potential strategies for improvement of circularity, the alkaline batteries offer a suitable product for integrative circularity-environment assessment intended to highlight advantages and limitations of assessment methods and potential for their joint use. Two recycling routes provide separate baseline scenarios of current practices in battery management which are extended to consider other improvements in battery design and management (detailed in Section 2.2.3.).

## *2.2.2. Assessment methodology*

Assessment of battery scenarios is carried out using LCA and MCI. The LCA provides a quantitative evaluation of battery impacts across all stages in the life cycle of batteries including all the inputs and outputs of the energy, materials use and waste emissions arising in the manufacture, use and disposal of the product, by investigating their potential contribution to several environmental impact categories. Consistent with ISO 14040 and 14044 standards, modeling is carried out through four phases: goal and scope, life cycle inventory, impact assessment, and interpretation (ISO-14040 2006; ISO-14044 2006). In contrast, the calculation of MCI is based on material flow efficiencies to reach a single integrative score. Data, calculation procedures and results for MCI are incorporated in the four phases of LCA framework: calculation procedure is outlined in goal and scope phase, data for quantification is detailed in life cycle inventory phase, and the values presented as part of the life cycle impact assessment phase.

### **2.2.2.1. Goal definition**

The primary objective of this study is to evaluate the trade-offs between environmental impacts and circularity of different battery management scenarios in order to improve methods combining and contribute to the efforts of circularity evaluation at the product level. Prospective findings are of interest to both industry and academia that require adequate methods to operationalize the circular economy and broaden the scope of assessment beyond environmental analysis.

### **2.2.2.2. System boundaries and functional unit**

All evaluated scenarios include manufacturing, use, and disposal of batteries. Also included are impacts of transportation between these stages involving battery purchase, spent battery collection, and their delivery to a recycling facility. The batteries are credited for avoided impacts of the production of virgin materials as a result of materials recovery through recycling. Batteries are manufactured mostly using primary (virgin) resources, and for some scenarios portion of secondary steel and zinc derived through the closed-loop (i.e., from recycled batteries) or open-loop, from the recycling of galvanized steel. Allocation of burdens among byproducts of recycling is made using 50-50 approach in which case impacts of manufacture of the upstream product providing secondary material to batteries and recycling is shared between upstream product and materials recovered through recycling in 50/50 ratio. The cut-off to the system boundaries includes capital goods such as infrastructure for recycling, buildings and transport vehicles, and plastic containers for collection and transportation of spent batteries. Some of these impacts are assumed to be insignificant or not known as battery production is not location-specific or locations are not known.

A sensitivity analysis is carried out to determine the robustness of the result to assumptions made to system boundary related to sourcing and modeling of secondary material used for battery manufacture and recycling. Two new scenarios are described in the following section.

The functional unit for comparison between battery life-cycle alternatives was to supply 1Wh of electricity. Considering the average battery capacity of 2450mAh operating at 1.5V AA battery, approximately 0.27 of single AA battery is needed to supply 1Wh of electricity.

Boundaries pertaining to the evaluation of MCI require less detailed description as the indicator is mostly calculated based on material flow rates. One important assumption made here was that  $E_r$  parameter pertaining to recycling efficiency (see Table 1), is determined based on material flows of base elements rather than the weight of recovery of materials in their oxidized or wet state. This was necessary for a calculation to be viably applied as batteries mass and volume increases to approximately 20% during their use and disposal (Olivetti, Gregory, and Kirchain 2011). If buffered weight was used instead, MCI value could theoretically surpass the maximum value of 1.

Limitations to data include a lack of specificity to certain material and waste datasets and occasional data gaps and assumptions. Average data from other publications were used to model manufacture, retail, transportation and collection whereas data for recycling is mostly based on thermodynamic estimates and information on transportation distances were not specified in a source report and therefore excluded or approximated. Given limitations to data are appropriate given the goal of this study which is used for demonstrative purpose.

#### 2.2.2.3. Product systems: alkaline batteries

Each scenario represents alkaline batteries of both AA and AAA type that are produced, used and disposed of in a specific manner for which several alternative battery life cycle routes comprising common and prospective battery management strategies are differentiated. Alkaline batteries consist of steel casing, brass connectors, zinc electrode, manganese electrolyte, copper connectors, and PVC and paper separator. As a baseline we consider battery life cycles comprising average technology and one of the two recycling routes, that can further incorporate one of “improvement strategies”. Baseline scenarios are explained in detail and each of improvement strategies is explained in reference to the baseline scenarios. Flow diagram of battery product systems adopting two recycling routes is depicted in Figure 2-1. All data considering recycling routes are obtained from an industry report commissioned by Raw Material Company (McLean Consulting 2014)

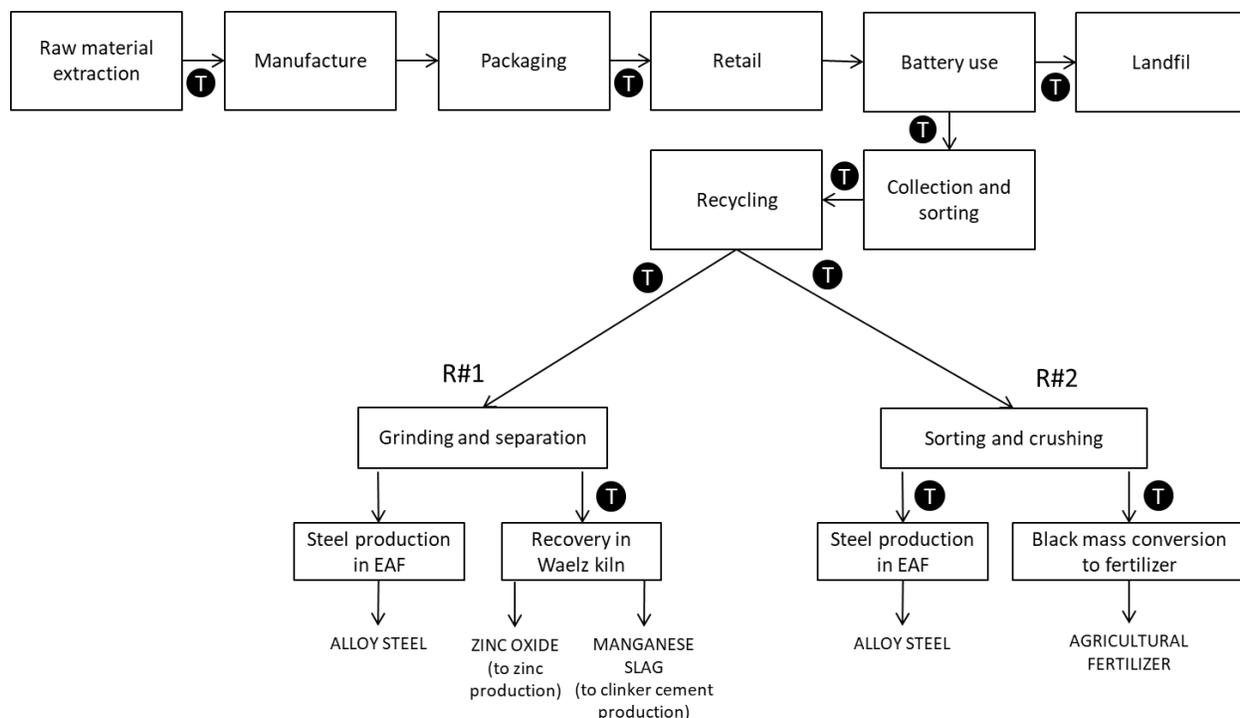


Figure 2-1. Battery life cycle including production and two alternative recycling routes. Flow diagram also includes the transportation (T-symbols next to the arrows), and byproducts of recycling.

1. **Baseline R#1.** This scenario represents the current design and management of batteries. Batteries are assumed to perform at an average capacity of 2450mAh per AA-type alkaline battery, manufacturing is carried out using exclusively virgin feedstock and end-of-life collection rates are consistent with current collection rates in Ontario (50%). For this scenario, it is assumed that 100% of collected batteries are recycled considering route #1 (R#1) which recovers materials at 83% efficiency, and corresponds to conventional practice involving three companies: Call2Recycle that enables collection, Inmetco that carries out the initial processing and sorting of batteries, and Horsehead zinc smelter that recovers material from the electrodes and electrolyte. In route #1 the spent batteries are first shredded to separate steel and other components. Steel is fed in electric arc furnace (EAF) to produce low alloyed steel. Remaining components: wrapper, copper connectors and electrodes, are fed to Waelz kiln to recover zinc-oxide and slag. Zinc-oxide is purified to zinc and slag reused as a substitute to a clinker cement. 50% of batteries not collected are assumed to be landfilled.
2. **Baseline R#2.** Manufacture, collection and disposal rates, and battery capacity in this scenario are analogous to baseline R#1 whereas the recycling is carried out by route #2 (R#2). This route comprises more refined mechanical separation to segregate steel,

wrapper, brass and electrodes. Steel casings are sent to steel smelter where they are recovered in EAF. Wrapper and brass are sent to energy recovery facility but recycler - Raw Material Company is not certain of their recovery and does not claim credits for potentially recovered brass and energy. Electrode material (black mass) consisting of zinc and manganese oxides, water, potassium hydroxide and carbon, is converted into agricultural fertilizer. The overall recovery efficiency of base elements is 83%.

Improvement scenarios apply to each of the recycling routes and include:

1. **Recycled content.** This scenario entails that 10% of the primary material in the manufacture of batteries is substituted by secondary material. Mainly, 50% of zinc and 50% of steel that constitute steel casing and electrode are substituted by secondary steel and zinc sourced either from the closed or open-loop recycling, as both metals could be recycled to high purity from batteries themselves, or sourced as recovered from other processes. Both closed-loop and open-loop scenarios are possible for batteries by R#1 while only the open-loop route is applicable to batteries by R#2 since recovered materials are used dissipative in agriculture. The closed-loop scenario is practically possible considering current recovery rates for R#1, i.e., the recovery rates of steel and zinc at the assumed collection rates and recycling technology, are sufficient to address the demand for 10% of recycled material proposed in this scenario.

The recycled content rate of 10% is higher than current rates of recycled content use for the manufacture of batteries but lower than predicted future rates by some of the producers. Energizer® EcoAdvanced™ alkaline batteries currently contain 4% recycled content, and the company predicts that this might increase to up to 40% in the next six years.

2. **Improved collection.** In this prospective scenario we assume that additional 10% of batteries are collected. Collection in Ontario is carried out through bi-annual curbside collection and designated public drop of facilities. The present collection rates of around 50% (Stewardship Ontario 2016), are targeted for further increase with introduction of bans and extended producer responsibility programs in Ontario and the rest of Canada (Giroux 2014; SagisEPR 2013; Stewardship Ontario 2009; Turner and Nugent 2015).
3. **Improved utility.** By this scenario, we assume designing batteries with longer shelf life or higher capacity that would lead to an increase of discharge current delivered over the lifetime. Although, the amount of energy that could be supplied through batteries dependent on multiple factors including the initial capacity, self-discharge, application, and environmental conditions, there are notable differences between how much energy can be supplied among the battery brands. For this scenario, we assume that the capacity

of batteries is increased by 20% in comparison to the industrial average, and capacity assumed for the baseline scenario.

4. **Maximum circularity.** This scenario is an integrative scenario of all three improvement strategies: batteries are produced with 10% recycled content, have 20% higher energy capacity in comparison the industry's average, and 60% of spent batteries are collected. This scenario is critical to our approach for visualizing MCI values in a normalized manner to be compared with normalized results of indicator values in LCA.

Two additional scenarios are considered for sensitivity analysis applicable to route #1. We compare how impacts and circularity is influenced when recycled content is delivered in a closed loop, as a possible alternative to default open-loop system. Additionally, we investigate how circularity and environmental impacts are affected by the decision to exclude credits for avoided burden production of clinker cement. In the latter case, two close-loop recycled content scenarios are compared, the default one and one excluding credits for clinker cement that affects impacts both upstream (in production of batteries) and end-of-life (credits allocated to batteries for recovered material).

#### 2.2.2.4. Impact assessment methods

Classification and characterization of material and energy inputs and waste outputs throughout the life cycle of batteries are carried out using OpenLCA v1.5.0 software and utilizing the cumulative energy demand (CED) method and ReCiPe endpoint (H) impact assessment method: human health, ecosystem, and resources (Goedkoop et al. 2008).

Calculation of MCI is detailed in the EMF report and consists of calculating three main parameters: the amount of virgin feedstock (V), unrecoverable waste (W), and utility (F) calculated by accounting several sub-parameters, as shown in Table 2-1. MCI of a product can be calculated as a weighted sum of MCI scores of each of the product components, or MCI directly calculated for the product. The per-component analysis allows to implement economic weighting to component MCIs to derive the final value. However, if a product's MCI is calculated based on mass fractions of components, MCI can be computed directly for the whole product with the same results (Lonca et al. 2018). For calculation of MCI of batteries, we pursue the direct approach.

MCI scores have been communicated both as their absolute values, increase or decrease from those values in relative terms, and also relative to the maximum circularity of batteries (i.e., normalized to maximum circularity scenario). Latter representation allows to interpret MCI scores and be able to compare them with impact categories and indicators.

The values of product circularity are inverted to those of environmental impacts (i.e., the higher graph for circularity indicate a more desirable outcome whereas high impacts of end-point impact categories and indicators are less desirable).

This approach allows better visualisation and perception of trade-offs between MCI and LCA impact categories and indicators.

Table 2-1.

Parameters considered and calculations in the assessment of MCI

<b><i>Parameters to be measured</i></b>	<b><i>Symbols and calculation</i></b>
Mass of the product	$M$
Fraction of feedstock derived from recycled resources	$Fr$
Fraction of feedstock derived from reused resources	$Fu$
<b>TOTAL MASS OF VIRGIN MATERIAL</b>	$V = M(1 - Fr - Fu)$
Fraction of the mass of the product being collected for recycling at EOL	$Cr$
Fraction of the mass of the product going into component reuse	$Cu$
Amount of waste going to landfill or energy recovery	$Wo = M(1 - Cr - Cu)$
Efficiency of the recycling process at product's EOL	$Ec$
Quantity of waste generated in the recycling process	$Wc = M(1 - Ec)$
Waste generated to produce recycled content used as feedstock	$Wf = M(1 - Ef) \frac{Fr}{Ef}$
Efficiency of the recycling process used to produce the recycled feedstock	$Ef$
<b>TOTAL MASS OF UNRECOVERABLE WASTE</b>	$W = Wo + \frac{Wf + Wc}{2}$
Product's lifetime	$L$
Average industries' product lifetime	$Lav$
Product's intensity of use	$U$
Average industries' product intensity of use	$Uav$
<b>UTILITY</b>	$X = \left(\frac{L}{Lav}\right)\left(\frac{U}{Uav}\right)$
<b>MATERIAL CIRCULARITY INDICATOR</b>	$MCI = 1 - \left(\frac{V + W}{2M + \frac{Wf - Wc}{2}}\right) 0.9X$

### 2.2.3. Life cycle inventory

#### 2.2.3.1. Data for LCA model

All foreground data for battery manufacture, packaging, retail, collection and recycling were adopted from previous publications and report commissioned for one of the recyclers. All

background data of material, energy and waste burdens are derived from the Ecoinvent v3.3. Background data for chemicals is assumed as average global, and electricity is modeled for the province of Ontario. Disposal of metallurgical waste is assumed as average slag from a specific industry, EAF slag, or average non-hazardous waste submitted for incineration.

Data for battery manufacture, packaging and retail, including transportation of batteries and materials are adopted from previous work (Dolci et al. 2016). According to this inventory, the batteries are manufactured by universal approach combining hydro and hard metallurgy. The electrolyte is produced in a solution by adding acid in solution of zinc and manganese. Steel casings are hot rolled and copper connectors produced by extrusion. Packaging assumes cardboard boxes that are used 0.48g per functional unit.

The data for secondary steel and zinc used as used for battery manufacture in the recycled content and maximum circularity scenarios is assumed from galvanized steel scrap recycling. The recovery process, detailed and adopted from previous work presumes steel recovery in EAF and zinc recovery from baghouse dust in Waltz kiln (Viklund-White 2000). 20kg of dust is assumed to be generated per tonne of steel (Antrekowitsch et al. 2014), electricity consumption for shredding of steel scrap is assumed 150kWh/tonne of scrap steel, and materials and energy for the production of steel from scrap in EAF obtained from the report (Stubbles 2000). Recycling of galvanized steel is carried out at 95% efficiency.

We assume that all batteries in Ontario are either collected as part of the municipal waste stream or collected separately and recycled. Impacts for the collection of batteries includes transportation from the collection point to sorting facility and then to recyclers. Batteries collected as part of municipal waste stream are transported directly to the landfill. Distances for both legs of transportation are adopted from previous work on batteries (Olivetti et al. 2011). The first leg includes the collection of batteries with 20t municipal solid waste truck driving the distance of 35 km at 60% capacity to sorting facility or 270km to the landfill at 80% capacity (Olivetti et al. 2011). Batteries collected for recycling are further transported from a sorting facility to the recyclers. For route #1 this includes 500km distance to Philadelphia, and for route #2, 300km to Port Colborne.

Data for recycling of batteries was derived from a thermodynamic report prepared by McLean consultancy (McLean Consulting 2014). The main material flows detailed in the report are shown in Table 2-2. Zinc oxide as a byproduct from route #1 undergoes an additional step of reduction to elementary zinc according to inventory found in (Viklund-White 2000).

Table 2-2.

Inputs and outputs of two recycling routes considered in this study.

Alkaline battery disposal, route #1			Alkaline battery disposal, route #2		
INPUTS	Quantity	Units	INPUTS	Quantity	Units
Spent batteries	1000.0	kg	Spent batteries	1000.0	kg
Electricity (for separation)	38.0	kWh	Electricity (for separation)	38.0	kWh
Electricity (for EAF)	144.0	kWh	Electricity (for EAF)	144.0	kWh
Coke (for Waelz kiln)	88.0	kg	Sulphuric acid	271.0	kg
Oxygen (for Waelz kiln)	124.4	kg			
OUTPUTS			OUTPUTS		
Slag (from EAF)	2.6	kg	Carbon dioxide	16.6	kg
Carbon monoxide	268.8	kg	Slag (from EAF)	2.6	kg
Carbon dioxide	80.8	kg	Baghouse dust (to waste)	1.5	kg
Chlorine	12.4	kg	Steel low-alloyed (avoided)	190.1	kg
Baghouse dust (to waste)	31.2	kg	Zinc sulfate (avoided)	147.7	kg
Mn-Cu-Fe slag (avoided)	271.3	kg	Manganese sulfate (avoided)	229.0	kg
Crude zinc oxide (avoided)	231.3	kg	Potassium oxide (avoided)	31.6	kg
Steel low-alloyed (avoided)	190.1	kg	Zinc oxide (avoided)	149.0	kg
Baghouse dust (ZnO for recovery)	5.2	kg	Manganese oxide (avoided)	239.0	kg

### 2.2.3.2. Data for calculation of MCI

Data for calculation of MCI, with the exception of calculation of utility (F), is based on material and process efficiency rates. Data does not incorporate flows of energy and auxiliary emissions such as transportation and capital goods. Material flows for the recycling of batteries was made based on material flows of base elements as shown in Table 2-3. Battery treatment routes achieve the same efficiencies of 83%. The data used for each parameter for calculation of MCI for each battery scenario is shown in Table 2-4.

Table 2-3.

Per element recovery rates of R#1 and R#2 per 1000 kg of spent alkaline batteries.

	Units	Materials in alkaline battery	Route #1	Route #2	Route #1 (no slag reuse)
Zinc (electrode)	kg	190	185	178	185
Manganese	kg	250	250	250	0
Iron	kg	190	185	184	184
Nickel	kg	4	4	4	4
Zinc (in casing)	kg	0	0	0	0
Potassium	kg	26	0	26	0

Carbon	kg	36	0	0	0
Copper	kg	20	20	0	0
Zinc (in brass)	kg	10	0	0	0
PVC	kg	15	0	0	0
Nylon	kg	15	0	0	0
Paper	kg	15	0	0	0
Total	kg	772	644	641	372
Recovery rate	%		83	83	48

Table 2-4.

Material circularity indicator parameter values for baseline and improvement scenarios, including recycled content (close-loop) and no clinker credit which is used in the sensitivity analysis.

MCI parameters	Baseline	Recycled content. (CL)*	Recycled content (OL)	Improved collection	Improved technology	Maximum circularity	No clinker credit*
M	1000	1000	1000	1000	1000	1000	1000
Fr	0	0.10	0.10	0	0	0.10	0
Fu	0	0	0	0	0	0	0
Cr	0.5	0.5	0.5	0.6	0.5	0.6	0.5
Cu	0	0	0	0	0	0	0
Ec	0.83	0.83	0.95	0.83	0.48	0.83	0.48
Ef	0.83	0.83	0.83	0.83	0.83	0.83	0.48
L	1	1	1	1	1.2	1.2	1
Lav	1	1	1	1	1	1	1
U	1	1	1	1	1	1	1
Uav	1	1	1	1	1	1	1

\* - applies only to R#1

## 2.3. Results

### 2.3.1. Life cycle impact assessment

#### 2.3.1.1. Absolute circularity and environmental impact values

MCI values of batteries differentiated for two recycling routes, and related improvement scenarios are shown in Table 2-5, including also scenarios investigating the influence of byproduct characterization (i.e., no clinker credit), and closed-loop recycling, both applicable to the route #1 only.

It can be observed that MCI values for route #1 and route #2 are identical, due to the same recycling efficiencies, giving a value of 0.29 for the baseline scenario. For a single-strategy

improvement scenario, it can be observed that the highest value for MCI is achieved with the increased utility, followed closely by improvement in recycled content, and lastly, collection rates. Maximum circularity by incorporating all these strategies equals 0.48, which is 0.19 increase of indicator value or a relative increase of 65% in comparison to the baseline scenario.

Table 2-5.

Absolute values of end-point categories, cumulative energy demand and MCI for baseline, improvement scenarios, and scenarios used in sensitivity analysis, for R#1 and R#2.

		Units	Baseline	Recycled content (CL)	Recycled content (OL)	Improved collection	Improved technology	No clinker credit	Max circularity
Batteries by R#1	H.H.	DALY	1.04E-07	9.73E-08	1.06E-07	1.03E-07	8.65E-08	1.00E-07	8.68E-08
	Res.	\$	1.76E-02	1.73E-02	1.76E-02	1.75E-02	1.46E-02	1.73E-02	1.45E-02
	Ecosys.	spec.yr	3.81E-10	4.47E-10	3.89E-10	3.78E-10	3.16E-10	4.59E-10	3.21E-10
	CED	MJ	5.11E-01	5.13E-01	5.14E-01	5.14E-01	4.24E-01	5.17E-01	4.30E-01
	MCI	-	0.29	0.33	0.34	0.33	0.41	0.26	0.48
Batteries by R#2	H.H.	DALY	9.27E-08	-	9.43E-08	8.90E-08	7.69E-08	-	7.48E-08
	Res.	\$	9.94E-03	-	9.92E-03	8.37E-03	8.25E-03	-	6.66E-03
	Ecosys.	spec.yr	1.11E-10	-	1.19E-10	5.43E-11	9.25E-11	-	4.30E-11
	CED	MJ	3.94E-01	-	3.97E-01	3.73E-01	3.27E-01	-	3.09E-01
	MCI	-	0.29	-	0.34	0.33	0.41	-	0.48

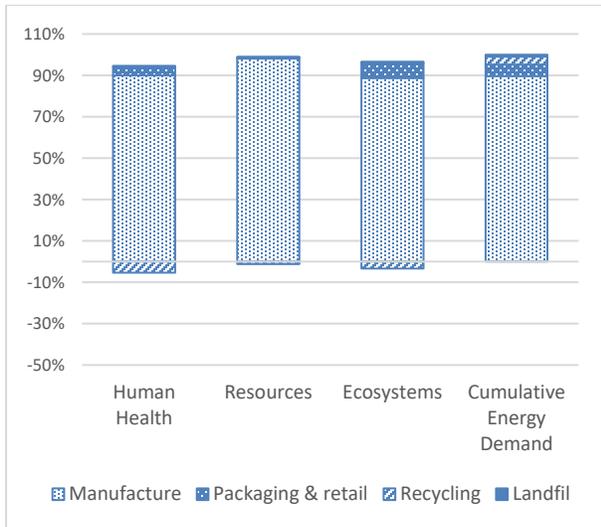
### 2.3.1.2. Baseline scenario comparison

Environmental impact contributions to product life cycle stages for two baseline routes are shown in Figure 2-2 (a, b), and their relative comparison is shown in Figure 2-3. It can be observed that the manufacture of batteries creates most of the impacts in the life cycle of batteries for both routes. For batteries by R#1, impacts of manufacture are followed by the small influence of packaging and retail, recycling, and the negligible contribution due to landfilling of batteries not collected for recycling. In comparison, the recycling stage in the life cycle of R#2 has a more substantial impact on the life cycle of batteries. Relative benefits of the recycling for R#2 baseline are in the range of 12-42% for three categories and indicator of cumulative energy demand, and nearly outweigh the impacts of manufacture for the end-point category of ecosystems.

Relative comparison between baseline scenarios (Figure 3) shows that baseline R#2 have significantly lower impacts than R#1. Impacts are lower by roughly 70% for ecosystems, 40% for resources and 12% for human health end-point categories. Clearly, the results of environmental impacts significantly diverge from MCI values that are equal for two scenarios (MCI=0.29), calculated based on the efficiency rates of their recycling routes that are both estimated at 83%.

Identical efficiencies of two recycling routes allow to highlight how influential is the nature of secondary material to either circularity or environmental impacts of recycling, but also on the other strategies that are affected by characteristics of secondary material. The choice of recycling route affects whether the collection is beneficial and whether materials can move in a closed or open loop. R#1 enables both open and closed-loop recycling, while the use of materials recovered in R#2 is limited to a specific secondary application involving fertilizer production.

a)



b)

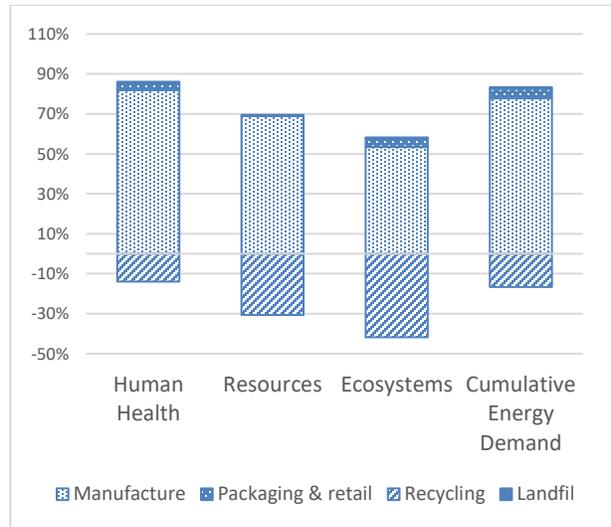


Figure 2-2. Contribution analysis for cradle-to-cradle life cycle of batteries for baseline routes: a) batteries by R#1, and b) batteries by R#2.

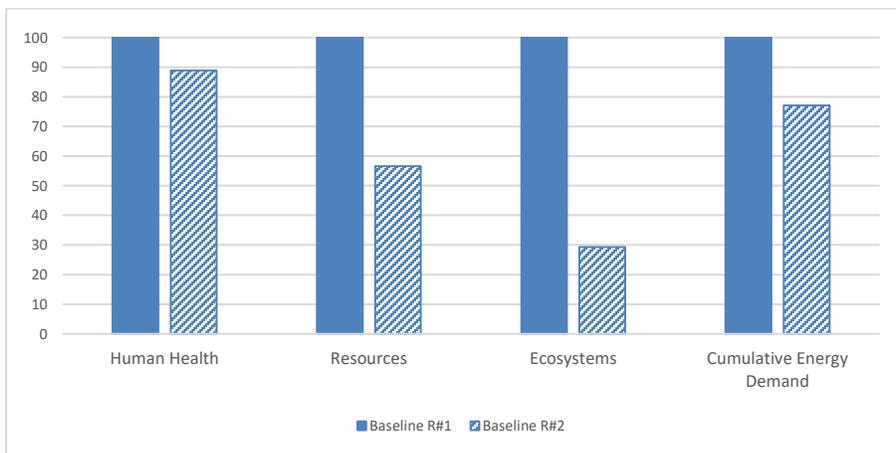
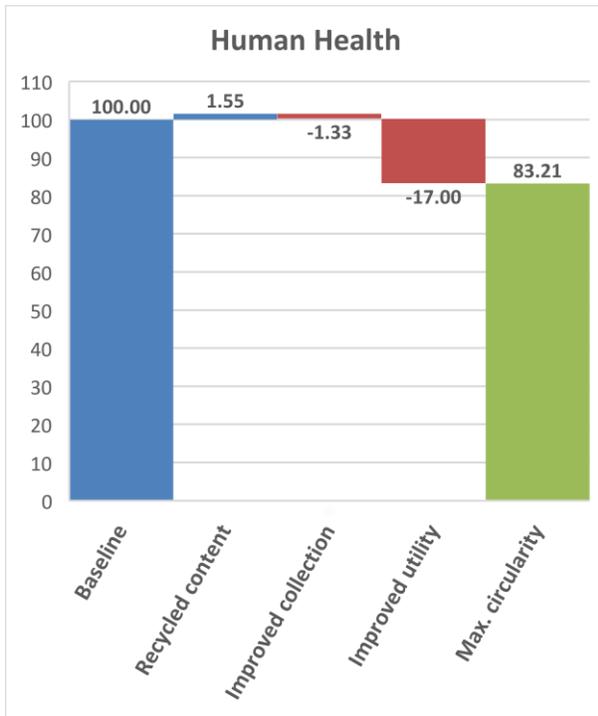


Figure 2-3. Normalized comparison of two baseline routes for end-point categories and cumulative energy demand.

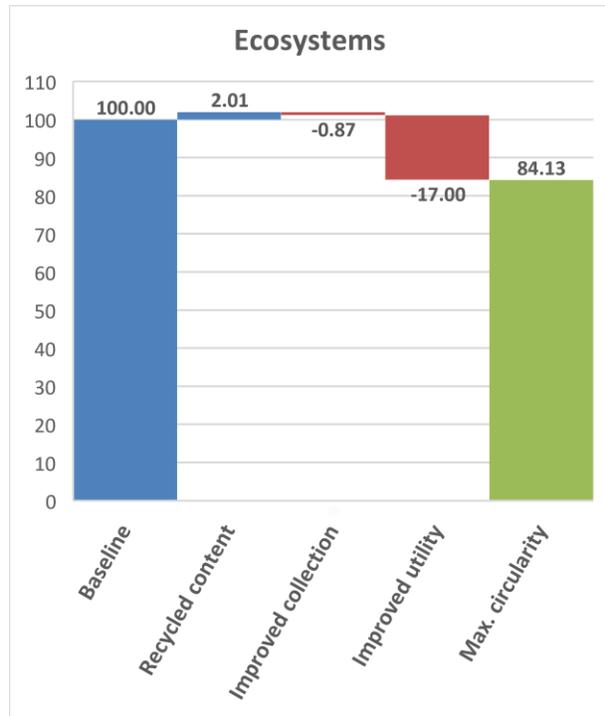
### 2.3.1.3. The relative contribution of improvement strategies

Evaluation of three improvement scenarios is made using normalized values for both LCA categories and indicators and MCI which allows to visualize the trade-offs between MCI and LCA categories and indicators. The environmental impacts are shown in Figure 2-4 (a-d) and Figure 2-5 (a-d), and MCI in Figure 2-6 (a-b). For categories and indicators of LCA, the comparison is made by showing increase or decrease in value of impacts for the scenarios relative to the baseline (at 100%), and MCI values are shown a relative increase of circularity to the maximum circularity scenario (at 100%). Waterfall charts are shown for recycled content derived through open-loop recycling.

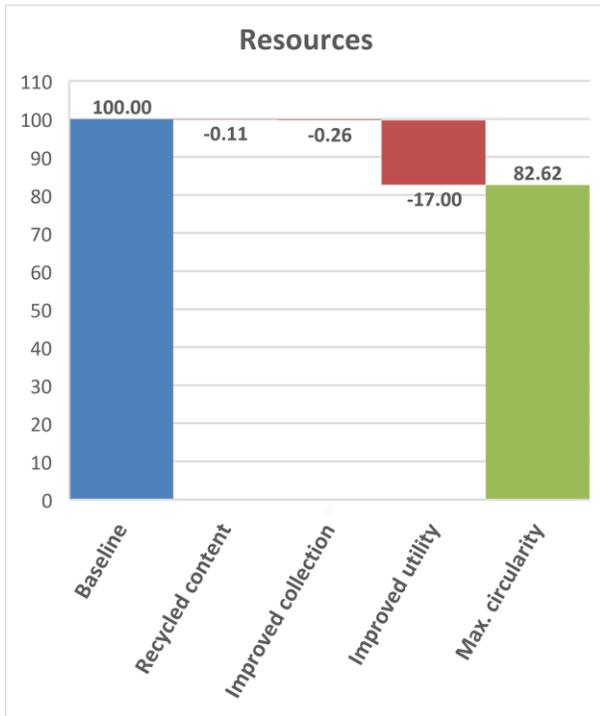
a)



b)



c)



d)

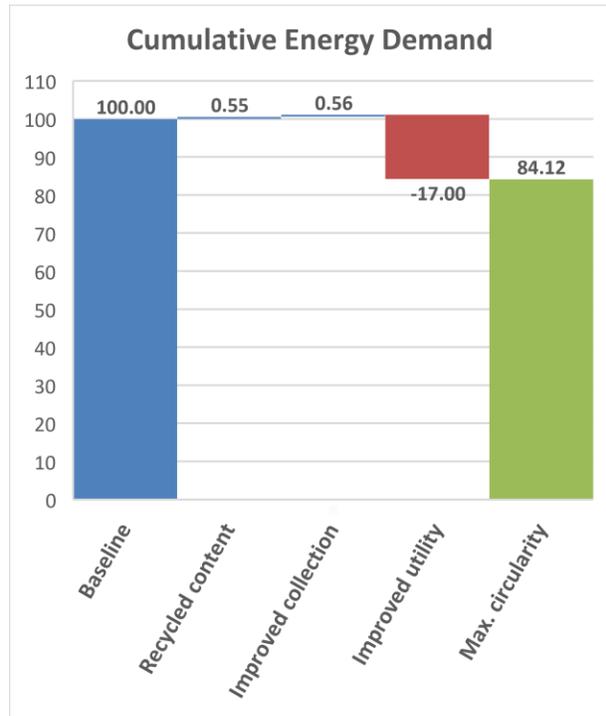
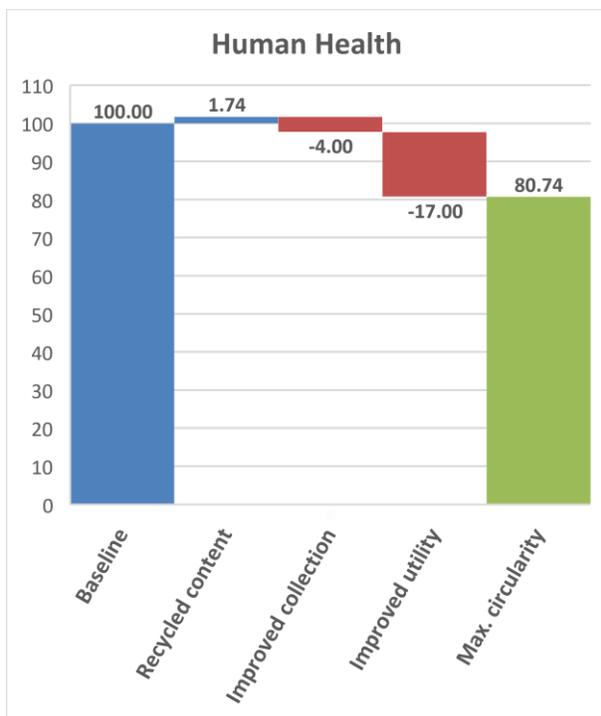
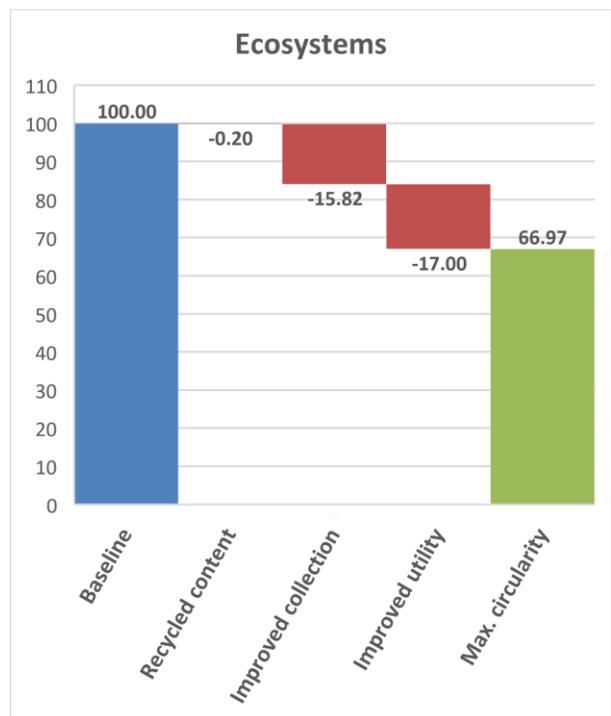


Figure 2-4 (a-d). The relative effect of improvement strategies on impact reduction in reference to the baseline for batteries by route #1.

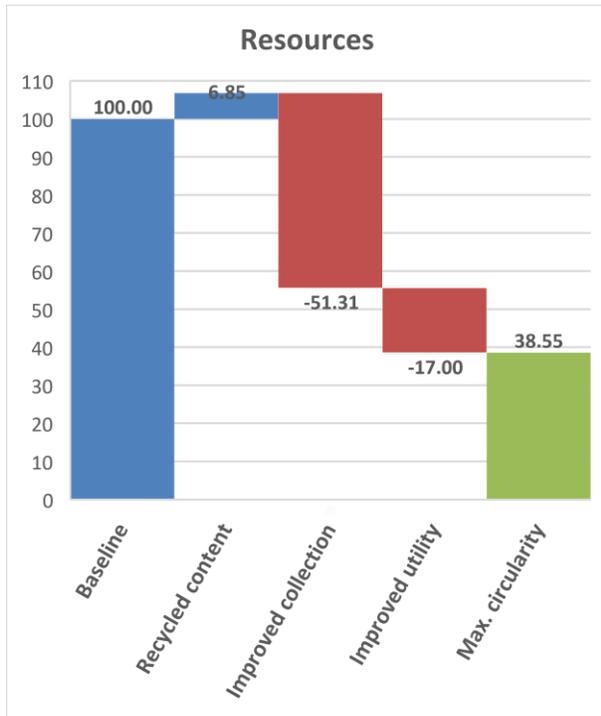
a)



b)



c)



d)

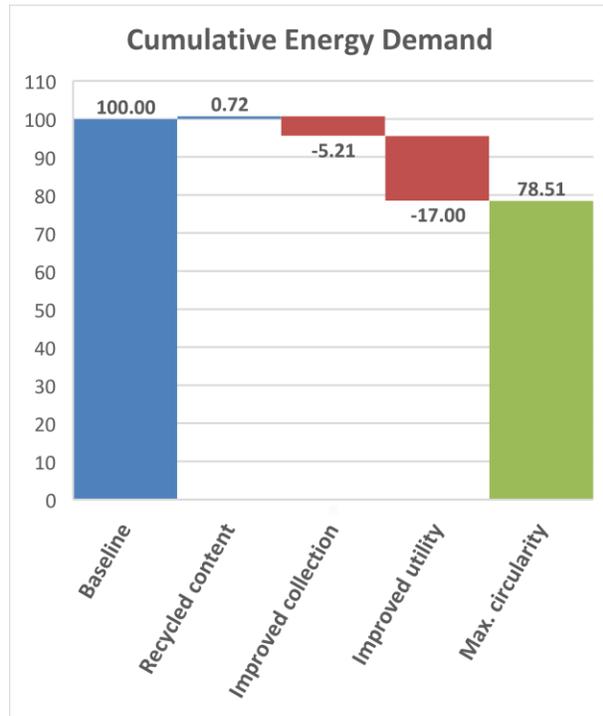
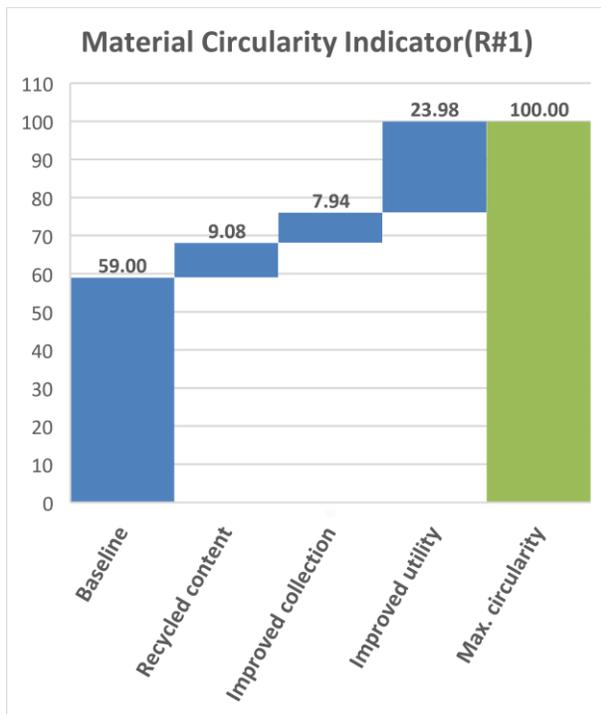


Figure 2-5 (a-d). The relative effect of improvement strategies on impact reduction in reference to the baseline for batteries by route #2.

a)



b)

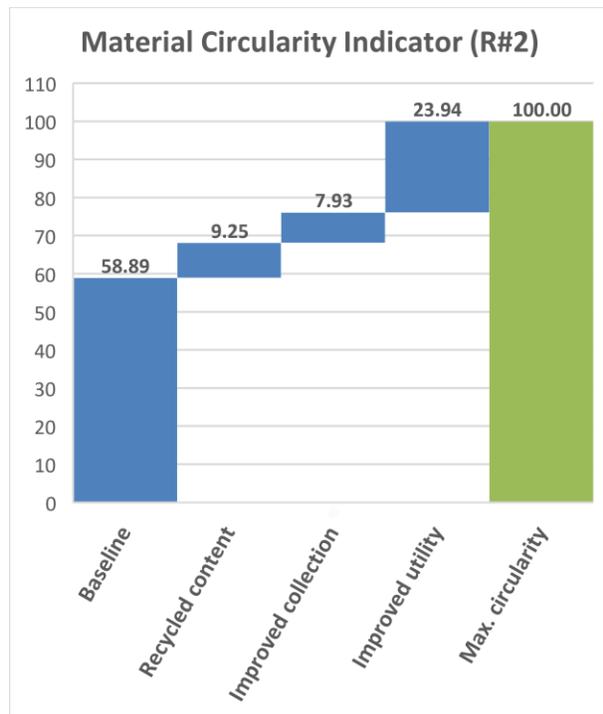


Figure 2-6 (a-b). The relative effect of improvement strategies to MCI-circularity increase in reference to the maximum circularity potential scenario (identical for R#1 and R#2).

Deriving from the values of both MCI and LCA categories and indicators, it appears that the majority of applied strategies reduce environmental impacts. However, it can be observed that only improvements in utility are uniformly complementary for two methods, and have similar values (17 for LCA in comparison to 24 for MCI). The values of other improvement scenarios for LCA and MCI do not significantly correlate in value, and sometimes in direction. Recycled content most significantly diverges for two methods. For the resources category, use of recycled content represents a small improvement in comparison to the baseline for route#1 (0.2%) whereas it negatively affects this category for route #2, creating more substantial trade-offs (6.85%). For both routes, MCI increases by around 9%. Collection of batteries in all cases reduces environmental impacts since recycling creates benefits for all impact categories and indicators. Although, complementarity is not reflected in value, with impact categories and indicators being vastly affected by choice of recycling route. Increase in circularity is relatively small for MCI (~8%) while for impact categories and indicators improvements due to recycling are negligible for R#1 (1-2%) while for R#2, impact reduction is substantial (4-51%).

### *2.3.2. Interpretation*

#### *2.3.2.1. Influence of byproduct characterization on indicator trade-offs*

Comparison between batteries incorporating recycled content in a close-loop and batteries with recycled content derived through open-loop (i.e., secondary zinc and steel from galvanized steel recycling) for R#1, is shown in Figure 2-7. Incorporating recycled content in battery manufacture through closed-loop recycling improves environmental impacts for human health by 7% and resources by 3%, but has increases impacts for ecosystems by 1%. The MCI values are similar for two scenarios with MCI of 0.35 for open-loop and 0.34 for closed-loop scenario. The difference of 0.01 or a relative increase of 3% in circularity, is the result of a slightly higher recovery rate for recycled material derived through open-loop (95% in comparison to 83% for a close-loop). In this case, although relative percent change is the same for the end-point category of resources and MCI, and overall median percentage change between impact categories and indicators is similar, MCI and LCA categories and indicators are different in direction with MCI showing preference to recycled content derived from open-loop recycling. These values are observed for 10% recycled content use and would further increase if more secondary material would be used in product manufacture. With such increases, the relative influence on human health category would clearly become quite substantial given the choice of secondary material origin. On the other hand, MCI is only concerned with how efficient is the recycling process to obtain recycled content, and indifferent to the material origin. We calculated that if the primary feedstock is

entirely substituted by secondary material owing recycling efficiency of 83%, MCI of batteries would equal 0.71, and if that efficiency was 95%, MCI would be 0.74, thus only slightly higher.

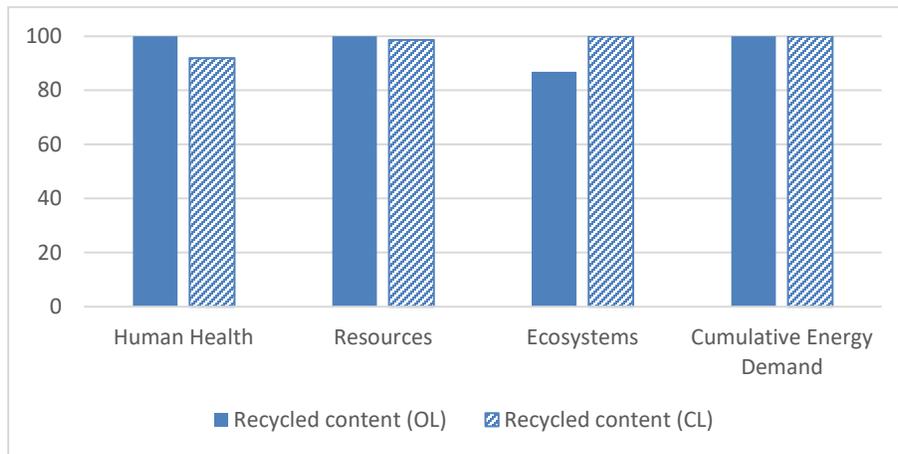


Figure 2-7. Comparison between close (CL) and open-loop (OL) recycled content scenarios for R#1.

Also, in relation to choices made to product system boundary, Figure 2-8 shows how LCA categories and indicators are impacted when the system is not credited for avoided production of clinker cement assumed as a substitute to manganese slag byproduct for route #1. This scenario reflects on the aspect that manganese reuse as clinker is a case of downcycling that has as a main aim a diversion of waste rather than motivated production.

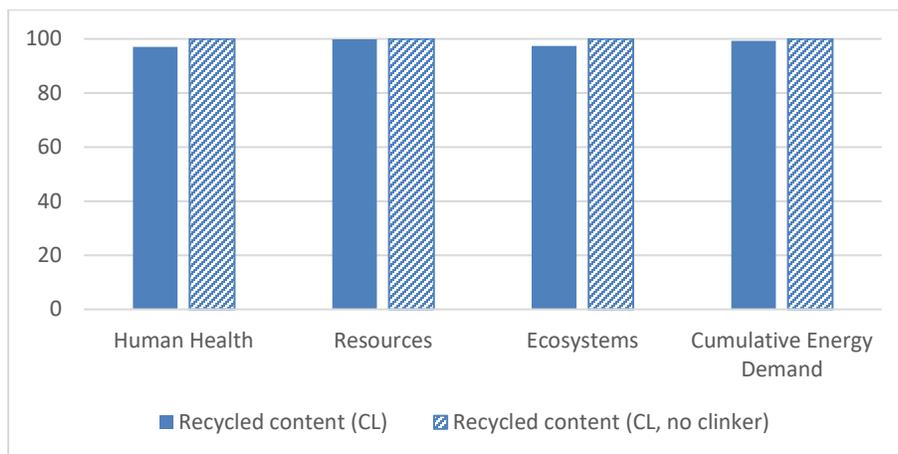


Figure 2-8. Comparison between recycled content close-loop scenario and recycled content close-loop scenario excluding clinker cement as a byproduct.

For impact categories and indicators, in line with the 50-50 allocation method, the exclusion of clinker as a byproduct increases the share of environmental burdens appropriated to the secondary zinc and steel used as recycled content. In this instance, the burdens for production

and recycling of batteries are now shared only between zinc and steel byproducts, and not a slag. On the other hand, for MCI, exclusion of clinker results in lower material flow efficiency rates and affects parameters of recycling efficiency ( $E_r$ ) and efficiency to produce recycled content ( $E_p$ ), which reduce from 0.83 to 0.48.

It is evident that the characterization of clinker only minimally affects LCA results while MCI indicator is more considerably affected. Environmental impacts decrease less than 3%, whereas MCI value decreases from 0.33 to 0.25 for the no-clinker scenario, which is a 30% decrease in relative value. Inconsistent responsiveness of indicator values to truncation of system boundaries affects the robustness of the dual analysis and undermines their joint use.

## 2.4. Discussion

Observed extent and consistency of the trade-offs between environmental and circularity performance renders two questions: should MCI adapt more alongside environmental choices, and how can MCI be adjusted or supported to improve their joint use (i.e., avoid significant trade-offs, and become more consistent with environmental categories and indicators under changing modeling assumptions)? Two questions are discussed, including also advantages and limitations of the new approach for interpretation of MCI value.

### *2.4.1. Opportunities and limitations of normalized MCI approach*

Representation of MCI scores shown in Figure 6 mirrors conventional representation of impact values in LCA allowing visualization of trade-offs between MCI and LCA categories and indicators and their combining. The approach rests on the assumption that multiple competing or complementary strategies are often available and needed to implement together in order to improve circularity (Blomsma and Brennan 2017; Kalverkamp and Young 2019). A sum of these multiple strategies serves as a yardstick to estimate how each strategy is individually valued in advancing product's circularity and assigns its value in range 0-100.

In addition to the visual and methodological appeal, an advantage of this approach is a representation of MCI score inherent to the context of technology and its practical limitations to close the loop of resources (i.e., advance on parameters quantified by MCI). MCI score normalized to its maximum circularity potential reflects on the notion that every product has different potential to achieve circularity, which is useful outlook if the progress for improving circularity among different products wants to be achieved at an equal footing. Such normalization of MCI value could be useful as a coefficient performance indicator to monitor implementation of the

circular economy adaptation across different product assortments and sectors within a company given different nature and level of challenges that different products face to improve circularity.

The need for normalization of MCI scores has been voiced previously (Niero and Kalbar 2019). In that instance, the authors normalize MCI scores to their Positive Ideal Solution and LCA categories to their Negative Ideal Solution and add another step of weighting to arrive at the single score. In the referred study, normalization is made against the best possible scenario for packaging, whereas maximum circularity value chosen as a reference in our study is a sum of strategies that could be applied at different life cycle stages. In the studies by Lonca et al. (2018) and Walker et al. (2018), MCI values are normalized and inverted to be compared with impacts in LCA (i.e., material linearity). The graphical representation adopted in Walker et al. (2018) bears some resemblance to the approach proposed in our study. However, similarly to Niero and Kalbar (2019) normalization is made in reference to the most impactful scenario.

In addition to argued advantages, it needs to be noted that normalized MCI value could not be used to compare different products (meant to fulfil the same function), in which case only absolute values are appropriate.

#### *2.4.2. Implications to MCI use and development*

Sizable trade-offs between environmental impacts and circularity for certain strategies in this and previous studies renders the question of whether MCI choices should try to adapt more alongside environmental lines. Although, an improvement in circularity should not be expected to accompany environmental improvements as this has been shown and argued earlier (Geyer et al. 2016; Humbert et al. 2009; Linder et al. 2017; Lonca et al. 2018), an instance of significant trade-offs for specific strategies might be an indication that circularity indicator needs further improvement, or the scope of its implementation has to be contextualized for technologies and evaluated strategies. In case of investigated strategies for batteries, only the strategy of improved utility entails consistent improvements for both environmental impacts and circularity while the strategies of recycling and improved collection are only meaningful if recycling creates benefits and its benefits are sizeable in comparison with impacts from other life cycle stages. An adaptation of circularity indicator more in line with environmental choices could prevent that circularity strategy is dismissed and also ensure, that in case LCA is not carried out, the circularity strategy does not result in significant trade-offs to the environment.

Another potential aspect for MCI development, affecting its use with LCA, relates to a lack of consistency between MCI and LCA categories and indicators with decisions to change or truncate product system boundaries. As we have shown, the decision to exclude byproducts had

significant impacts on MCI but a minor influence on environmental categories and indicators suggesting that the use of recycled content, as well as recycling, are poorly supported. This shortcoming is likely exacerbated for circularity assessment of already manufactured products in which case circularity improvements are limited to end-of-life strategies. In contrast, characterization of primary material quality in MCI is conveniently addressed through a measurement of the product utility. Such inconsistency could be further affected by other choices in LCA modeling such as the selection of different allocation methods or changes in value of the product utility that does not affect value of MCI in a linear fashion as this is case for LCA, i.e., small increase in product utility makes a steep increase of MCI and plateaus after reaching certain value.

To potentially enhance consistency and also ameliorate trade-offs between categories and indicators of LCA and MCI, the circularity assessment with MCI could be complemented with other circularity indicators, providing certain hierarchy among competing alternatives for end-of-life treatment, or revising  $E_r$  parameter calculation to incorporate the recycling gradient. Multiple circularity indicators have been developed exclusively to support end-of-life management practices and secondary material quality that could be used to complement MCI given some additional data of end-of-life procedures, material pricing, or market potential (Huysman et al. 2017; Linder et al. 2017; Di Maio et al. 2017; Di Maio and Rem 2015; Moraga et al. 2019; Park and Chertow 2014; Vanegas et al. 2018; Zink et al. 2016). Circularity indicator proposed by Huysman et al. (2017) allows quality characterization using proxy of exergy, while a proxy of price in a value ratio between input product and secondary (recycled or reused) material is also frequently used (Linder et al. 2017; Di Maio and Rem 2015). More indirectly, material quality can be grasped looking at easiness of disassembly that evaluates product fractions separation (Vanegas et al. 2018), or looking at the reuse potential of material on the market (Park and Chertow 2014; Zink et al. 2016)

The role of secondary material quality evaluation for added consistency to coupling is likely an important one, but not an exhaustive. Recovery of materials of sufficiently high quality is critical in reaching higher recovery rates in circular economy (Pauliuk 2018). Finally, it is important to bear in mind that MCI, and any other circularity indicator or combinations thereof, need to strike a good balance between simplicity and accuracy, so as to remain attractive to industry practitioners to be used alongside other performance indicators in a straightforward and feasible manner. In the current study, although we briefly discuss how MCI could shape to better support coupling and environmental choices, we don't discuss the broader norms of circular economy and what measurement of circularity should entail. For that, the readers should refer to other works (Elia et al. 2017; Linder et al. 2017; Potting et al. 2017; Saidani et al. 2017).

## 2.5. Conclusions

Assessment of the products by combining LCA and MCI could help identify circularity strategies that are both environmentally sound and minimize the use of resources. The challenges to combining arise due to methodological differences to calculate indicator values, different valuation scales and means to resolve the trade-offs between MCI and LCA categories and indicators. To confront these challenges, we apply two methods to several strategies for the design and management of alkaline batteries and observe the extent and consistency of indicator trade-offs. The comparison between MCI and LCA categories and indicators is made possible by normalizing MCI values, by identifying the maximum circularity potential of batteries and measure how each individual strategy fare in contributing to that maximum value. This approach improves visualization of trade-offs and allows to incorporate the context of technology and its potential to improve circularity, which could be useful for companies to manage their internal efforts in adaptation of circular economy. Deriving on the results, we pinpoint the strategies that are more viable to consider when applying MCI and advocate caution if MCI is used to inform recycling and use of recycled content in product manufacture to improve circularity. We conclude that MCI and LCA categories and indicators combining would improve with better characterization of secondary material quality losses and discuss how additional circularity indicators could be used to improve on that aspect.

## Chapter 3: Life cycle assessment of emerging Ni-Co hydroxide charge storage electrodes: impacts of graphene oxide and synthesis routes <sup>2</sup>

### Abstract

Decoupling energy supply from fossil fuels through electrification and sustainable energy management requires efficient and environmentally low-impact energy storage technologies. Potential candidates are charge storage electrodes that combine nickel and cobalt hydroxides with reduced graphene oxide (rGO) designed to achieve high-energy, high-power density and long cycling lifetimes. An early eco-efficiency analysis of these electrodes seeks to examine the impacts of materials and processes used in the synthesis, specifically while focusing on the use of rGO. The emerging electrodes synthesized by means of electrodeposition, are further compared with electrodes obtained by an alternative synthesis route involving co-precipitation. Life cycle assessment (LCA) method was applied to compare a baseline nickel-cobalt hydroxide electrode (NCED), the focal electrode integrating rGO (NCED-rGO), and the benchmark co-precipitated electrode (NCCP), for delivering the charge of 1000mAh. Contribution analysis reveals that the main environmental hotspots in the synthesis of the NCED-rGO are the use of electricity for potentiostat, ethanol for cleaning, and rGO. Results of comparison show significantly better performance of NCED-rGO in comparison to NCED across all impact categories, suggesting that improved functionalities by addition of rGO outweigh added impacts of the use of material itself. NCED-rGO is more impactful than NCCP except for the categories and indicators of cumulative energy demand, climate change, and fossil depletion. To produce a functional equivalent for the three electrodes, total cumulative energy use was estimated to be 78 W·h for NCED, 25 W·h for NCED-rGO, and 35 W·h for NCCP. Sensitivity analysis explores the significance of GO efficiency uptake on the relative comparison with NCCP, and potential impact of GO in process effluent on the category of freshwater ecotoxicity. Scenario analysis further shows relative performance of the electrodes at the range of alternative functional parameters of current density and lifetime. Lastly, the environmental performance of NCED-rGO electrodes is discussed in regard to technology readiness level and opportunities for design improvements.

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<sup>2</sup> A version of this chapter was accepted for publication in the journal of RSC Advances as: Edis Glogic, Alberto Adán-Más, Guido Sonnemann, Maria de Fatima Montemor, Liliane Guerlou-Demourgues, Steven B. Young (2019). Life cycle assessment of emerging Ni-Co hydroxide charge storage electrodes: impacts of graphene oxide and synthesis routes. Journal of RSC Advances, vol. 9, no 33, p. 18853-18862.

### 3.1. Introduction

Energy storage technologies are considered essential in the pursuit of sustainable energy use, especially their foreseen role in decarbonizing transportation sector and the expansion of renewable energy infrastructure. To meet increasingly diversified demand of these and other applications, efforts in materials science have been directed on developing energy storage systems with improved functionality and low environmental impacts. Desired properties are expected for storage systems with high-energy density, high-power density and long cycling lifetimes of their integrating electrodes (Chae, Zhou, and Chen 2012; Conway 1991; Rydh 2003).

The state-of-art electrodes could be developed by combining metal oxides and hydroxides with carbon-based materials (Nguyen et al. 2017; Prioteasa et al. 2015; Simon and Gogotsi 2008; Y. Wang, Song, and Xia 2016), which are currently pursued in positive electrodes for batteries and hybrid-supercapacitors based on nickel and cobalt (Ni-Co) hydroxides fabricated by means of potentiostatic electrodeposition (Adán-Más et al. 2017). Enhanced electrochemical performance of the Ni-Co hydroxide electrodes has been observed with the addition of reduced graphene oxide (rGO). The addition of rGO improves the capacity of the electrodes and acts as a conductive matrix that accommodates strain in the charge-discharge process leading to longer lifetimes (Adán-Más et al. 2017). Conveniently, the reduction of GO to rGO is facilitated through the process of electrodeposition itself, avoiding the energy-intensive step of chemical reduction or other analogous routes, which would otherwise be necessary.

Considering their emerging nature, quantification of the environmental impacts of novel electrodes has not been previously attempted. Such an early inclusion of environmental performance consideration in the design of emerging electrodes could ensure that the potential impacts in electrode synthesis are known and can be minimized in the future design and process optimization. Several studies, that constitute closest available literature, include analysis of several emerging cathode composites that only loosely resemble some of the materials used for the fabrication of novel electrodes (Deng et al. 2018; Peters et al. 2016). Similarly, lack of science-based environmental impact analysis applies to the exploration of a new trend of combining metal-hydroxides with carbon-based materials such as rGO.

To address these gaps, the present study employs the life cycle assessment (LCA) method to establish resource, ecosystem and human health impacts in the synthesis of emerging electrodes, implications of addition of GO to metal hydroxide matrix, and performance of electrodes as they are compared with existing alternatives. The analysis identifies how impacts of the electrodes can be improved, and what functional application leads to optimized environmental performance

with the aim to support these electrodes to take a positive role in sustainable energy management.

## 3.2. Materials and methods

The LCA study is carried out through four phases pertaining to requirements and recommendations of the International Standards Association, including (i) goal and scope, (ii) life cycle inventory, (iii) life cycle impact assessment, and (iv) life cycle interpretation (ISO-14040 2006; ISO-14044 2006).

The goal and scope phase, detailed in the current material and methods section, outlines the study purpose, the boundaries of the modeled product systems (i.e., electrodes), the function and functional unit used as a reference for comparison, and select impact category indicators used for characterization of environmental impacts.

### 3.2.1. Goal and scope

#### 3.2.1.1. Goal definition

The goal of this study is to identify environmental hotspots in the synthesis of Ni-Co electrodes integrating rGO, and determine if the addition of rGO improves eco-efficiency when compared with baseline electrodeposited Ni-Co hydroxide electrode and Ni-Co hydroxide electrodes obtained through co-precipitation (an alternative fabrication route).

Given this goal, the analysis aims to improve and determine the eco-efficient status of the rGO integrating electrode and, as a broader objective, shed light on environmental implications of newly adopted practice in the materials science of combining carbon-based materials and metal hydroxides and oxides. The findings of this study are meant to contribute to the material design and to support future research on energy storage materials, thus informing material scientists and technology developers working on energy storage.

#### 3.2.1.2. Function and functional unit

The function of the electrode is defined by its ability to store energy over its effective lifetime, characterized by two parameters: (1) the electrode's capacity, representing an ability of the material to store charge given applied current, and (2) the number of charge-discharge cycles that can be carried out before capacity reduces due to structural degradation, chemical parasitic transformations or other ageing phenomena that occur during cycling. Capacity fade is commonly tolerated up to 20-30%, or in other words, when 70-80% of the initial electrode capacity is maintained (Chaari et al. 2011; Murray and Hayes 2015; Saxena et al. 2015). The functional unit

(FU) used to compare electrodes is generating discharge current of 1000 mA h over the lifetime of the electrode, at a current density of 1 A·g<sup>-1</sup> considering a maximum capacity fade of 20%.

$$FU = X \cdot \frac{\sum_{n=1}^{n=n_{EOL}} \left\{ \frac{\sum_{i=1}^{i=n} C_i}{n} \right\}}{n_{EOL}}$$

where  $C_i$  is the capacity of the material after  $i$  charge-discharge cycles in mA·h·g<sup>-1</sup> (electrode's capacity after the first cycle),  $n$  is the total number of cycles at a given point during the charge-discharge cycling test,  $n_{EOL}$  is the number of cycles to reach end-of-life capacity fade, and  $X$  is the mass of electrode's active layer, expressed in grams.

### 3.2.1.3. Product systems: NCED and NCED-rGO

The charge storage electrodes investigated in this study are nickel-cobalt hydroxides deposited on top of conductive stainless-steel substrate. The two electrodes are synthesized by means of potentiostatic electrodeposition. First product system is the baseline electrode NCED (Ni-Co-Electrodeposited), with chemical formulae  $\alpha\text{-Ni}_{0.33}\text{Co}_{0.66}(\text{OH})_2 \cdot (\text{CO}_3^{2-}, 2 \cdot \text{NO}_3^-) 0.66(\text{H}_2\text{O})_{0.5}$  and second, the analogous composite in which reduced graphene oxide is added: NCED-rGO, with chemical formulae  $\alpha\text{-Ni}_{0.33}\text{Co}_{0.66}(\text{OH})_2 \cdot (\text{CO}_3^{2-}, 2 \cdot \text{NO}_3^-) 0.66(\text{H}_2\text{O})_{0.5}/\text{rGO}$ . Derivation of the formulae is available in Supplementary Information (SI), Table S3-1.

Synthesis of the two electrodes and their characterization has been depicted in Figure 3-1 and detailed elsewhere (Adán-Más et al. 2017). The NCED electrode was prepared by applying a 10 second pulsed potential between -0.9 V and -1.2 V to the working electrode (stainless steel AISI 304) and using a counter electrode of platinum submerged in an aqueous electrolyte containing nickel and cobalt nitrate hexahydrates with a concentration of 3 mM and 6 mM respectively, and a saturated calomel electrode as the reference electrode. The deposition rate was approximately 1.5 µg per cm<sup>2</sup> per minute, assuming linearity (Streinz et al. 1995), which can be manipulated to achieve different layer thicknesses. The electrodeposited electrode material was subsequently washed with water and ethanol to remove impurities and to facilitate drying. The NCED-rGO electrode was prepared similarly, with the exception of the substitution of the aqueous electrolyte by a graphene oxide (GO) aqueous suspension at the concentration of 1 g·L<sup>-1</sup> and subsequent addition to electrodeposition bath along with the nickel and cobalt salts. Prior to its addition, GO was ultrasonicated for 30 min to ensure stable dispersion of GO flakes.

Electrochemical properties have been measured at several current densities including 1 A·g<sup>-1</sup>, 4 A·g<sup>-1</sup> and 10 A·g<sup>-1</sup> and after reaching capacity fade of 20% and 30%. The cycling stability and the consequent evaluation of capacity fade was assessed at the current density of 10 A·g<sup>-1</sup> by applying continuous charge-discharge during 5000 cycles in the 0.45 V to -0.2 V potential range. Cycling

stability was assumed to be equivalent for other current densities, as similar degradation phenomena occur.

At the applied current density of  $1 \text{ A} \cdot \text{g}^{-1}$ , the capacity of NCED electrode is  $30 \text{ mA} \cdot \text{h} \cdot \text{g}^{-1}$  and NCED-rGO is  $96 \text{ mA} \cdot \text{h} \cdot \text{g}^{-1}$ . For NCED the capacity fade of a 20% is reached after 972 cycles, and NCED-rGO after 1676 cycles.

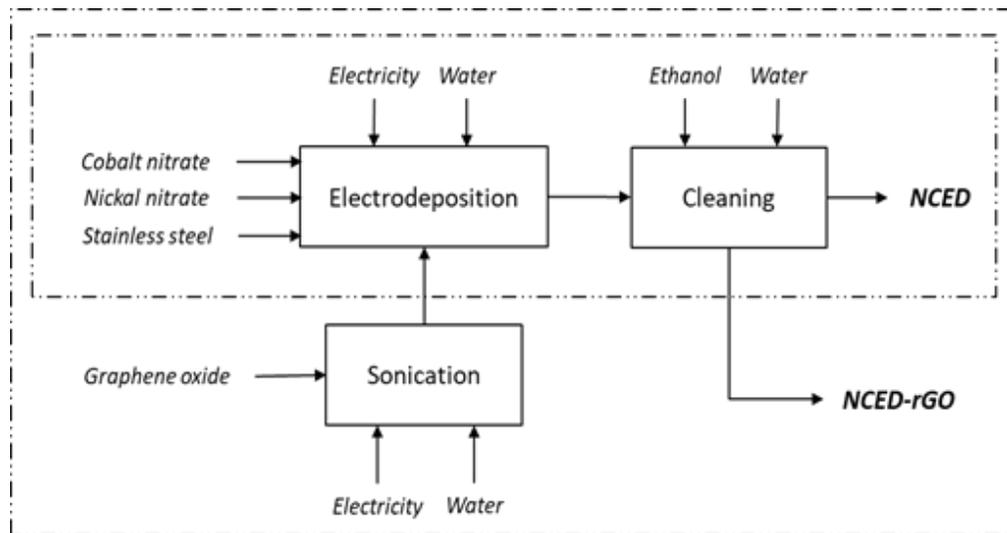


Figure 3-1. Process flowchart for the synthesis of NCED and NCED-rGO

#### 3.2.1.4. Product system: NCCP

More mature and resource-optimized Ni-Co hydroxide electrode involving the synthesis of active material by co-precipitation followed by physical deposition onto a stainless-steel substrate was used as a benchmark for comparison with NCGOED-rGO electrode. The material, noted here as NCCP (Ni-Co Co-Precipitated), with chemical formulae  $\alpha\text{-Ni}_{0.33}\text{Co}_{0.66}(\text{OH})_2 \cdot (\text{CO}_3^{2-}, 2 \cdot \text{NO}_3^-)_{0.66}(\text{H}_2\text{O})_{0.5}$ , is prepared in several process steps as depicted in Figure 3-2, and detailed elsewhere (Faure, Delmas, and Willmann 1991a, 1991b). Co-precipitation is carried out in 2M sodium hydroxide solution, containing 1.5 mL of hydrogen peroxide, that is slowly added to a solution containing 5 g of nickel nitrate hexahydrate and 10 g of cobalt nitrate hexahydrate (1:2 molar ratio). The mixture is stirred for 48 h and washed under centrifugation (4000 rpm, 10 minutes) six times with water and two with ethanol to reach stable pH. The precipitate is dried in the oven for 24 h at 40 °C. The material, in powder form, is then mixed with polytetrafluoroethylene (PTFE) and carbon black (at 80:5:15 mass ratio, respectively), using ethanol as solvent, and pressure-printed on a stainless-steel grid to produce the electrode. The capacity of this electrode is  $121 \text{ mA} \cdot \text{h} \cdot \text{g}^{-1}$  at a current density of  $1 \text{ A} \cdot \text{g}^{-1}$ , and reaches a capacity fade of 20% after 1006 charge-discharge cycles.

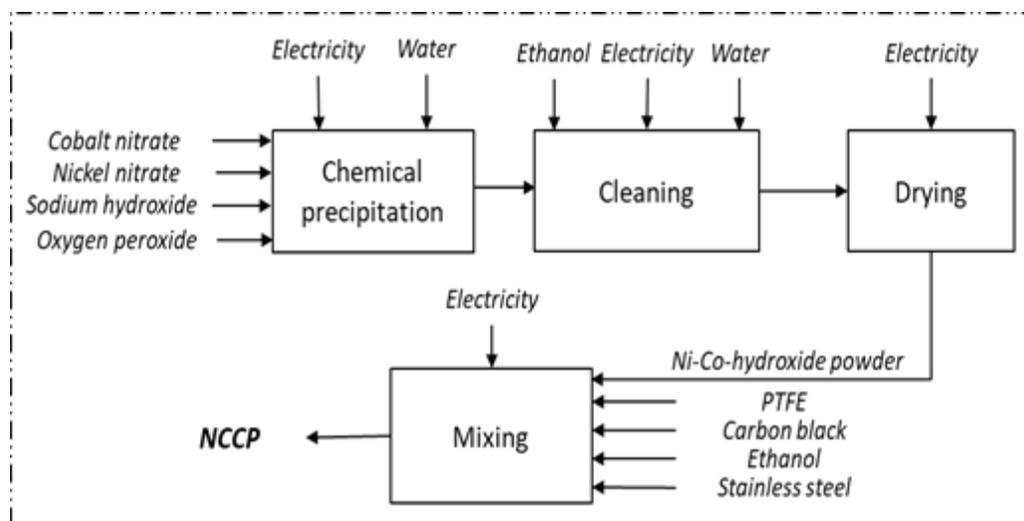


Figure 3-2. Process flowchart for the synthesis of NCCP

### 3.2.1.5. System boundaries

Cradle-to-product gate analysis was carried out. Boundaries of the analyzed product system include the production and manufacture of the electrodes, and do not quantify impacts arising in the use and disposal of the electrodes, which could have been modeled only in consideration to the entire energy-storage device. Cut-off criteria also apply to capital goods such as laboratory equipment, machinery, buildings and transportation vehicles used to carry out the processes and manage materials. Impacts of capital goods are assumed to be negligible considering low use of abrasive chemicals and high temperatures. In either case, these impacts would be almost identical for the two electrodeposited electrodes that employ similar processing steps. Capital goods also include the counter and reference electrodes that are used in electrodeposition, since they are not consumed in the process and could be reused without losses and deterioration.

Wastewater effluents from potentiostat, co-precipitation and washing stages are excluded for the baseline comparison and only potential toxicity impacts of GO to the freshwater systems quantified and investigated as a separate scenario. The small amounts of cobalt and nickel present in the effluent, in similar concentrations for all three electrodes, are assumed to be treated on site and precipitated prior to disposal to the sewage system or landfill. The literature suggests a high degree of removal of cobalt (up to 90%) and nickel (up to 80%) from the effluents (Fu and Wang 2011; Kurniawan et al. 2006). The reagents that would be potentially used to precipitate these pollutants are assumed to be small and uncertain, as electrolytic effluent is mixed with the wastewater from other experiments before it is treated. GO in effluent could be precipitated to a large degree using adsorption (Sun et al. 2018), floc-flotation (Chen and Li 2018), coagulation (Duan et al. 2017; J. Wang et al. 2016), or photo-degradation (Zhang et al. 2016). However, if the specific type of treatment is not applied to the process effluent before its disposal

to the conventional sewage system, the portion of GO could be eventually left untreated and enter natural water systems. GO in freshwater is associated with toxicity-related effects (Ou et al. 2016).

Classification and characterization of environmental impacts is carried out using the indicator of cumulative energy demand and nine impact assessment categories of ReCiPe Midpoint (H) method including: climate change, ionizing radiation, metal depletion, fossil depletion, water depletion, terrestrial ecotoxicity, freshwater ecotoxicity, marine ecotoxicity, and human toxicity (Goedkoop et al. 2008). GO impacts to freshwater ecotoxicity was investigated using USEtox method (Rosenbaum et al. 2008), as needed characterization factors and reference units of toxicity impacts were developed specifically for this method. Toxicity impacts in USEtox are expressed in the unit of potentially affected fraction of species (PAF) per cubic meter per day per kilogram emitted, i.e.,  $\text{PAF}\cdot\text{m}^3\cdot\text{day}\cdot\text{kg}^{-1}$ , in comparison with ReCiPe using kg 1,4-dichlorobenzene (1,4-DB) equivalents (Deng et al. 2017). The modeling was carried out using OpenLCA v1.5 software. Direct material inputs in the synthesis of the electrodes are either observed experimentally, scaled-up in reference to industrial practice or optimal use of laboratory equipment. Value assumptions are based on the literature, direct measurements, and expert opinion. Synthesis of the electrodes is modeled as a foreground system while impact-profiles of reagent materials are sourced from the Ecoinvent v3.3 database. Input quantities, related assumptions, and background data sources for each material in the foreground system are described in a subsequent LCI section. Input quantities of each material are shown per 1g of active material. Material inputs per quantity of active materials corresponding to the functional unit and naming of background data sourced from the Ecoinvent database are provided in SI, Table S3-2 and S3-3. Background datasets of electricity inputs and transportation were selected for an average European context, while for all chemicals an average global production was assumed.

In addition to sensitivity analysis applied to system boundaries to investigate potential toxicity effect of GO in the effluent, a sensitivity analysis is also carried out to address the inventory assumption related to an efficiency rate of GO use in the manufacture of NCED-rGO. The GO uptake of 80% was considered as a scaled-up scenario. The scaled-up NCED-rGO is compared with NCCP only.

Scenario analysis was further carried out to compare electrodes in consideration to different operational parameters of applied current density and electrodes' lifetimes. In comparison to the baseline scenario, in which electrodes are compared at  $1 \text{ A}\cdot\text{g}^{-1}$  current density and capacity fade of 20%, a comparison was also carried out at current densities of  $4 \text{ A}\cdot\text{g}^{-1}$  and  $10 \text{ A}\cdot\text{g}^{-1}$  and a capacity fade of 30%. In total, five scenarios are investigated. Performance values of the electrodes

(capacity and number of charge-discharge cycles) at these alternative current densities and capacity fade criteria are detailed in SI, Table S3-4.

### 3.3. Results

The results section encompasses the remaining three phases in LCA: the inventory, impact assessment, and interpretation phase. The inventory phase details data sources, assumptions and quantities for all the materials and energy inputs, which are then classified and characterized among environmental impact categories, in the subsequent impact assessment phase. In interpretation phase, main findings and modeling assumptions related to functional unit and GO use, are investigated through scenario and sensitivity analysis.

#### 3.3.1. *Life cycle inventory*

##### 3.3.1.1. Cobalt and nickel nitrates

The quantities of cobalt and nickel nitrate salts used during the fabrication of electrodeposited electrodes (NCED and NCED-rGO) are calculated assuming an uptake efficiency of 95%. These are scaled-up efficiencies readily achieved in industrial settings. Cobalt and nickel nitrate salts are modeled as obtained in direct oxidation from metallic cobalt in reaction with nitric acid (Grayson, Kirk, and Othmer 1996). 0.83 g of nickel-nitrate-hexahydrate and 1.661 g of cobalt-nitrate-hexahydrate were required to produce 1g of active material for fabrication of NCED, and 0.79 g and 1.58 g for fabrication of NCED-rGO electrode, respectively.

Use efficiency of cobalt in nickel salts in co-precipitation was calculated to be 83%. 0.76 g of nickel-nitrate-hexahydrate and 1.54 g of cobalt-nitrate-hexahydrate are required to produce 1g of active material for NCCP.

##### 3.3.1.2. Graphene oxide solution

Inputs of GO for fabrication of NCED-rGO had to be established on the basis of limited knowledge of GO behavior during electrodeposition, specifically, the efficiency of GO-use in electrodeposition and the concentration of the GO in the final electrode. It is approximated that rGO constitutes 5% of the NCED-rGO electrode which comes as an additional mass in electrodeposition in comparison to NCED, holding all other process material-inputs equal. This assumption is supported by the slight difference in weight between two materials. However precise measurement is difficult due to the possible presence of surface pollutants, non-homogeneous nature of the dispersion and limitations of the techniques currently used to quantify weight percentage (i.e., energy dispersive X-ray spectroscopy and X-ray photoelectron spectroscopy).

GO is used in excess in laboratory practice and only around 1% is utilized in the preparation of electrode. However, unlike the metal salts, the electrodeposition of GO is an emerging practice and the efficiency rates of deposition could not be emulated from existing industry practices and current scientific literature. Although, considering the non-ionic mechanism of GO deposition, and low reactivity of rGO, it is likely that this efficiency would be lower than for metals. Consequently, we made a conservative assumption in which GO use-efficiency is set at 5% for the default comparison, and uptake of 80% has been investigated as a potential scale-up scenario. Use efficiency will, among other, depend if the electrodeposition of GO can be established as a continuous and semi-continuous production, as opposed to batch-scale production carried out in a laboratory.

The inventory for the production of GO was adapted from comparative LCA study of different GO production routes (Cossutta, McKechnie, and Pickering 2017), based on Bangal variant of the Hummer's method (Chen, Yan, and Bangal 2010). 1 g of GO or 1000 ml of 1% GO solution was assumed per 1 g of active electrode material.

Electricity use for sonication of GO was assumed for 110 W sonicator working for 30 min mixing 1 L solution of GO, equaling use of energy to 55 W·h per 1 g of GO.

#### 3.3.1.3. Stainless steel substrate

A stainless steel AINSI 304 is used as a substrate for all electrodes, given its low electrochemical signal and chemical stability, which are needed to evaluate the electrochemical response of the active material. The stainless-steel foil, with a thickness of 3 mm, is applied as a substrate and is used to deposit 0.05 g of active material per 1 cm<sup>2</sup> of active electrode material. 3.2 g of stainless steel is required per 1 g of active material.

#### 3.3.1.4. Transport of materials

Transportation of materials and process chemicals is assumed for average European transportation following the recommendation of Frischknecht et al. (2007). Accordingly, the distances of 600 km by train and 200 km by a 32 t lorry are taken for all the chemicals and 200 km by train and 100 km by 32 t lorry for stainless steel (Frischknecht et al. 2007).

#### 3.3.1.5. Electricity use

Potentiostat electricity is calculated for constant use of 75 W Gamry Interface 1000TM potentiostat, depositing an area of 100 cm<sup>2</sup> steel substrate, at -1.2 V which can allow deposition rates of 0.15 mg and 0.1575 mg of active material per min per cm<sup>2</sup> for NCED and NCED-rGO, respectively. Energy consumption was measured empirically resulting in energy use of 317 W·h for 1 g of active material for both NCED and NCED-rGO.

In co-precipitation, electricity was measured for 2280 W Sigma 4-16K Centrifuge operating for a total of 80 min at 4000 rpm. Electricity for drying was indicated for Mermert oven 5800 W, for 24h at 40°C and measured empirically. The energy for printing the mixture on a steel substrate is assumed at 5 t·cm<sup>2</sup>. Per gram of active material, the energy requirement for centrifuge was measured to be 15.15 W·h, for drying 106.8 W·h and the energy for printing was calculated to be 2.72 W·h.

#### 3.3.1.6. Cleaning agents, electrolyte and effluent treatment

Use of ethanol and water for washing and drying of electrodeposited electrodes corresponds to an experimental procedure. 3mL of ethanol and 9 mL of water are estimated for cleaning 1 cm<sup>2</sup> surface area of the electrode. For deposition of 1 g of active material, we estimate 62 ml of ethanol and 200 ml of water for NCED, and NCED-rGO.

Cleaning in co-precipitation was carried out under centrifugation using 602 ml of ethanol and 230 mL of water per gram of active material.

#### 3.3.1.7. Additional chemicals for the synthesis of NCCP

In addition to Ni-Co precipitation, fabrication of NCCP comprises additional mixing and pressing step that involves use of additional materials including polytetrafluoroethylene, carbon black, hydrogen peroxide, ethanol and sodium hydroxides. The inventory for production of polytetrafluoroethylene (PTFE) used as a binder is obtained from Jungbluth et al. (2012). 0.05 g of PTFE is used per gram of active material. Quantities of other reagents include 0.15 g of carbon black, 0.2 ml of hydrogen peroxide, 250 ml of ethanol, and 0.5 g of sodium hydroxide, for which background data was obtained from the Ecoinvent.

### 3.3.2. Life cycle impact assessment

#### 3.3.2.1. Contribution analysis of NCED-rGO

The relative contribution of the main processes and materials in the synthesis of NCED-rGO to environmental impact categories are shown in Figure 3-3. Application of GO and electricity for potentiostat appear to be the major contributors to the environmental impact categories. Impacts of GO application are generated from electricity for sonication and GO manufacture, and are mostly equally shared among each other for impact categories, with the exception of metal depletion category that is affected solely due to GO manufacture. Potential impacts of electricity are particularly high in the category of ionizing radiation due to use of nuclear electricity in Europe. Impacts of cleaning are the third most significant, with the particular contribution to the category of terrestrial ecotoxicity, fossil depletion and cumulative energy use. Consumption of distilled water for cleaning and potentiostat is the major contributor to impacts of terrestrial

ecotoxicity (45%) mostly due to impacts of road transportation. Ethylene in production of ethanol is responsible to high impacts of ethanol especially to category of fossil depletion (35%). Stainless steel substrate has high impact on marine and freshwater ecotoxicity and resource categories including water and metal depletion, due to use of ferrochromium and ferronickel used for alloying. Impacts of nickel and cobalt nitrates which are relatively low when compared to other reagents, contribute to toxicity categories and metal depletion. Impacts of transportation of foreground materials are negligible.

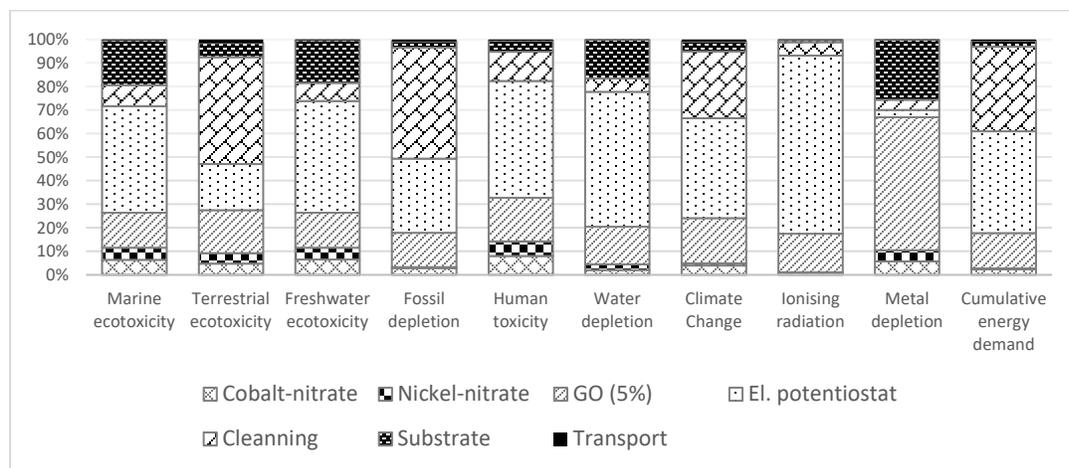


Figure 3-3. Environmental impact contributions of direct material use and emissions in the synthesis of NCED-rGO electrode

### 3.3.2.2. Comparison with NCED and NCCP

Normalized comparison between three electrodes is given in Figure 3-4 and 3-5, and absolute values detailed in SI, Table S3-5. In comparison to NCED, NCED-rGO electrode appears to have the lowest impacts in all investigated categories. Hence, it appears that significant improvement in electrochemical properties considerably outweighs added impacts from GO production. On average across all the impact categories, NCED-rGO generates 70% less impacts than NCED to reach the same discharge current.

The superior performance of NCED-rGO is not confirmed in reference to NCCP electrode. NCCP is better in most of the impact categories with on average 40% lower impacts. The exceptions are categories of fossil depletion, climate change and cumulative energy use that are driven by considerably higher use of ethanol and electricity in fabrication of NCCP.

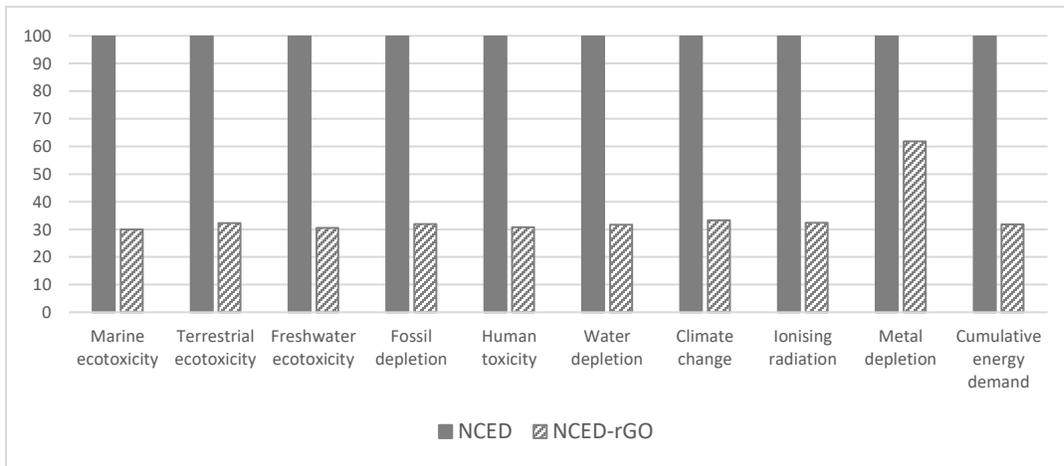


Figure 3-4. Normalized comparison between NCED and NCED-rGO

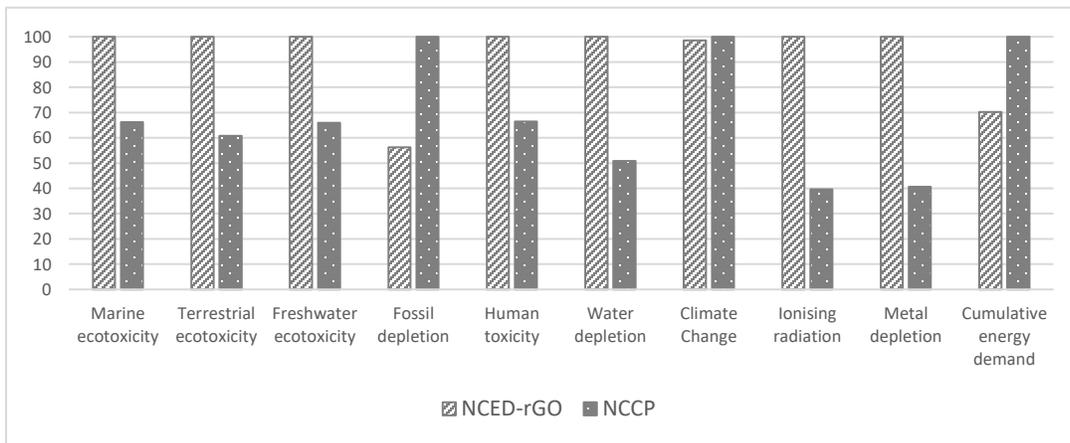


Figure 3-5. Normalized comparison between NCED-rGO and NCCP

### 3.3.3. Interpretation

#### 3.3.3.1. The potential impact of GO-rich effluent on freshwater ecotoxicity

Under the scenario that GO from effluent is not adequately treated and ends in freshwater systems, freshwater ecotoxicity impacts of NCED-rGO electrode would increase by 37% for default scenario or 8% for the scaled-up scenario (at GO use efficiency of 80%). As impacts of GO manufacture are 45% of total 15% that GO application adds to the net impacts in that category (remaining 55% are impact of electricity for sonication), the total impacts to category of

freshwater ecotoxicity would increase by 4% if GO would not be removed from the effluent before wastewater discharge. Absolute values of three electrodes and hypothetical scenario for NCED-rGO including impacts of GO to freshwater are given per FU in Table 3-1.

Table 3-1.

Relative comparison between electrodes for freshwater ecotoxicity including the scenario of untreated GO effluent (NCED-rGO+eff-GO). Unit CTUe [PAF m<sup>3</sup>.day.kg<sup>-1</sup>\_emitted] applies to all the values

	Freshwater ecotoxicity
NCED	7.68E-02
NCED-rGO	2.12E-02
NCCP	1.77E-02
NED-rGO + eff-GO	2.63E-02

### 3.3.3.2. Influence of increased rGO uptake on the relative comparison with NCCP

At more efficient GO use (uptake of 80%) two electrodes are comparable with clear preference for NCED-rGO in additional categories of terrestrial ecotoxicity and fossil depletion, in comparison to the baseline scenario. The comparison of the scaled-up scenario for NCED-rGO with NCCP electrode is given in Figure 3-6. GO use is one of the main environmental hotspots in the fabrication of NCED-rGO. On average GO production is responsible to 20% of the contribution in all impact categories and 70% of the impacts in metal depletion category. Overall, the significant influence of GO to toxicity categories results in notable shift in these categories at the scaled-up use. Contribution of GO to human toxicity, marine and freshwater ecotoxicity are divided between production of GO itself, and electricity use for sonication whereas terrestrial ecotoxicity is dominated by GO production (81%). Relative impacts to metal depletion that are entirely responsible by GO use and potassium permanganate (95%).

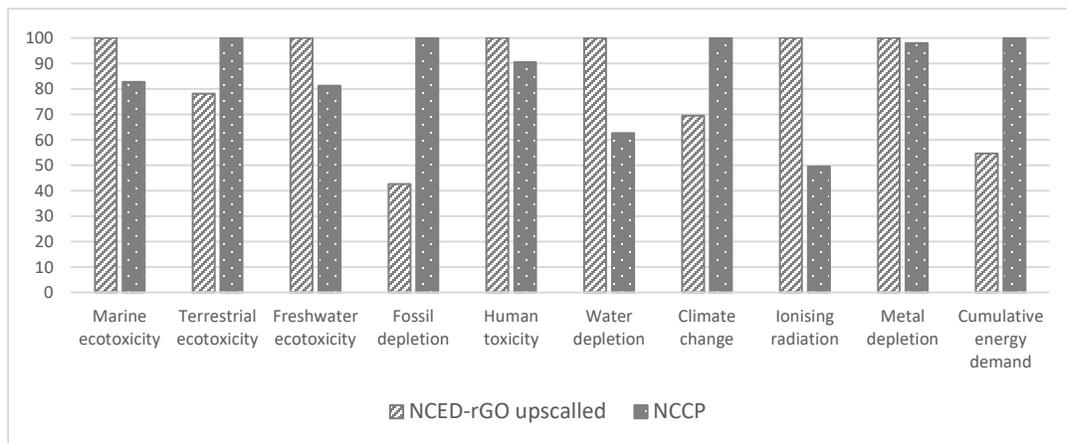


Figure 3-6. Normalized comparison between scaled-up scenario for NCED-rGO and NCCP

### 3.3.3.3. Effect of capacity fade and current density on relative performance of NCED-rGO electrode

The relative performance between NCED, NCCP and NCED-rGO electrode varies at different current densities and allowances for capacity fade. Table 3-2 shows relative performance of each NCED and NCCP in comparison with NCED-rGO electrode for baseline scenario corresponding to parameters taken for FU (S0), and additional scenarios (S-1, S-2, S-3, S-4, and S-5). Absolute values for electrode are given in Table S3-6 (a-c). It appears that NCED-rGO performs best for current density of  $1 \text{ A}\cdot\text{g}^{-1}$  considered for the baseline comparison, while it would entail slightly better performance at capacity fade of 30%. Overall, it compares similarly with NCED across additional scenarios. The preference for NCCP increases at higher current densities. Thus, the least favorable application of NCED-rGO appears at the current density of  $10 \text{ A}\cdot\text{g}^{-1}$ .

Table 3-2.

Relative impacts of NCED-rGO in comparison with NCED, and NCCP at different operational parameters of current density and capacity fade. Scenario abbreviation refer to combination of current density (CD) and capacity fade (CF): S-0 – CD 1 A·g<sup>-1</sup>, CF 20% (baseline); S-1 – CD 4 A·g<sup>-1</sup>, CF 20%; S-2 – CD 10 A·g<sup>-1</sup>, CF 20%; S-3 – CD 1 A·g<sup>-1</sup>, CF30%; S-4 – CD 4 A·g<sup>-1</sup>, CF 30%; S-5 – CD 10 A·g<sup>-1</sup>, CF 30%. Impacts of NCED-rGO are lower for percentage values preceded by the minus sign and are higher for positive values.

	Relative difference in comparison with NCED						Relative difference in comparison with NCCP					
	S-0	S-1	S-2	S-3	S-4	S-5	S-0	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	-70%	-60%	-70%	-76%	-66%	-73%	34%	61%	62%	29%	56%	59%
Terrestrial ecotoxicity	-68%	-58%	-68%	-75%	-64%	-72%	39%	63%	21%	34%	45%	49%
Freshwater ecotoxicity	-69%	-61%	-71%	-76%	-67%	-74%	34%	61%	18%	30%	57%	60%
Fossil depletion	-68%	-59%	-68%	-75%	-64%	-72%	-44%	-7%	0%	-51%	-22%	-16%
Human toxicity	-69%	-60%	-69%	-76%	-65%	-73%	34%	60%	19%	30%	56%	59%
Water depletion	-68%	-59%	-68%	-75%	-64%	-72%	49%	69%	23%	43%	65%	67%
Climate Change	-67%	-57%	-67%	-74%	-63%	-71%	-1%	39%	14%	-14%	26%	31%
Ionising radiation	-68%	-58%	-68%	-75%	-64%	-72%	60%	76%	25%	56%	71%	73%
Metal depletion	-38%	-20%	-39%	-52%	-31%	-46%	59%	75%	47%	56%	73%	75%
Cumulative energy demand	-68%	-59%	-68%	-75%	-64%	-72%	-30%	14%	20%	-39%	-2%	5%

## 3.4. Discussion

### 3.4.1. Opportunities and priorities for NCED-rGO design improvements

Results pinpointed several material and process hotspots that should be prioritized when considering future electrode design and scaling up processes, and while seeking improvements in specific impact categories. Although, this is not universally observed (Arvidsson and Molander 2017), the use of process reagents and energy that are modeled at the scale of laboratory equipment is expected to decrease with process optimization (Gavankar, Suh, et al. 2015; Gutowski et al. 2009). The impacts of materials that already assume industrial-scale efficiency could be further mitigated by identifying more eco-efficient substitutes or pursuing their recovery at the end-of-life. Here we discuss how impacts of some of the materials can be mitigated.

The impact of the stainless-steel substrate is relative to required active material thickness and a requirement for the thickness of the substrate itself, thus of concern if the aim is to create

electrodes with relatively thin deposits. To reduce impacts of the substrate, the stainless-steel could be compared with adequate alternatives which could include both metallic and non-metallic substrates (e.g., Dubal et al. 2014; Wu et al. 2006), or stainless-steel substrate used in a way that it could be recycled at end-of-life of the electrode (Frischknecht 2010).

The impacts of cobalt and nickel nitrates appear small but their share is likely to increase relative to a contribution to other electrode constituents since their use is already modeled at high efficiency. Reduction of impacts of cobalt and nickel salts can be targeted by manipulating their concentration taking their similar electrochemical properties in order to target reduction of specific impact categories. Relative to cobalt nitrate, nickel nitrate induces greater impacts in toxicity categories, while climate change and metal depletion categories are more impacted by cobalt nitrate. Such substitution is only appropriate under the condition that the functionality of the electrode is maintained.

The impact of GO could be mitigated with more efficient uptake of GO in the deposition process, and by using GO from more eco-efficient synthesis route. Increasing uptake would mean that less GO would be required to produce NCED-rGO electrode. Impacts of GO that are shown to be on average 20% would reduce to the low of 3-4% if GO would be utilized at the efficiency of 80%. Therefore, any efforts directed to better understand and improve GO deposition could significantly improve environmental impacts. For example, researchers and industry could examine the effect of increased deposition times and investigate if electrodeposition involving GO could be established as a continuous production.

The impact of GO appears to vary significantly depending on its manufacturing production route. Hence, sourcing or manufacturing of GO needs to be carefully selected and optimized if impacts of integrating electrode are to be minimized. The fabrication method adopted in the present study is based on chemical oxidation route (Chen et al. 2010), which rates favorably in comparison to approach by chemical vapor deposition and other variants of the Hummer's method (Cossutta et al. 2017). For example, the procedure initially considered, as described in Zaaba et al. (2017), was three times more impactful in comparison to chemical oxidation route we used for our model (Cossutta et al. 2017; Zaaba et al. 2017). Although, the authors report that impacts of adopted synthesis route could be potentially further reduced (by an average of additional 50%) if energy is used more efficiently, and hydrochloric acid is recovered and reused (Cossutta, McKechnie, and Pickering 2017).

### *3.4.2. Usefulness and limitations in view of emerging nature and technological maturity*

LCA of emerging technologies is often regarded as exploratory because findings are directional rather than definitive (Villares et al. 2017). In like manner, the findings of this study need to be interpreted in view of an emerging nature of studied electrodes, particularly the aspects of a less established functional requirement of the electrodes in comparison to the whole energy storage device, and technology readiness level (TRL).

Nonspecific functional requirement and flexible range of application of component-level emerging technology present the challenge to identify functional proxy in LCA. Representative functionality in our study is decided based on criteria of electrode's lifetime and operating current density, which ultimately correlate with the power requirements of the final device, a critical parameter in applications such as memory back-up applications or auxiliary power sources in small appliances such as laptops or mobile phones (Endo et al. 2001). However, since the final application of the electrode is not known, the parameters used for the functional unit (corresponding to baseline case), represents just one of the scenarios under which the electrode could be applied. Therefore, it is important to encompass other functional setups, by investigating multiple functional units or scenario analysis to reflect on possible alternatives as we have pursued in our study.

Observation of emerging technology's TRL is necessary to interpret the findings of the analysis and realize what is the potential of observed impacts and relative performance of electrodes to change as the technology matures. This is highly relevant when technologies of different TRLs are compared as they may own different potential that their impacts will change with future scaling-up and efforts to improve functionality (Gavankar, Suh, et al. 2015). In the current study, NCED-rGO appears at TRL level of four and NCCP at the level of five, which will likely make NCED-rGO more competitive as material develops. While comparing technologies at significantly different TRLs is not optimal (Troy et al. 2016), it is important to realize potential benefits to research and innovation of providing a certain benchmark to the development of new technology. Furthermore, different potentials of materials to undergo changes with increased scales means that the current relative contribution of different reagents, as shown in Figure 3-3, could further change. These potentials need to be considered when interpreting current results and prioritizing resource strategies to reduce the impacts of the electrodes.

With uncertainties pertaining to assessment of emerging technology, especially in regard to data, the primary aim of our study was to aid the design of the electrode and accommodate fair

comparison among competing electrodes of similar TRLs. Despite data and other limitations commonly claimed for assessment of emerging technologies (Hetherington et al. 2014; Wender and Seager 2011), incorporating eco-efficiency considerations to early-stage research has great potential to steer product design across more environmentally plausible lines (Villares et al. 2017; Wender et al. 2014). Specifically, analysis at an early stage can navigate most convenient application of technology at which technological and environmental performance are maximized, which should ultimately be the goal in materials engineering for energy storage. LCA can assist this process by selecting the most benign and convenient material while maintaining technical requisites.

In our study, we established that under favorable operational conditions, NCED-rGO has clear potential to be eco-efficient. Particularly, as such observation comes in addition to some of the other practical aspects inherent to electrodeposition fabrication method itself, that could add a positive impact on the environment. Electrodeposition enables morphology-controlled deposition enabling adhesion on various conductive substrates without the need of binders thus conferring certain flexibility to the technique (i.e., material thickness, surface geometry), involves few process steps required to produce the final electrode including the absence of thermal treatment of pressurized systems, and can be scaled to various capacities with ease (Schwarzacher 2006). These aspects result in shorter deposition times, safer working conditions and more durable use of process equipment, that could also positively affect environmental performance, but could not be captured in the present analysis. Nonetheless, this work reflects the need to increase capacity response and rate capability of electrodeposited Ni-Co hydroxide to produce more competitive results for this fabrication technique and NCED-rGO electrodes to be the most compelling choice. To this aim, strategies such as creating 3D hierarchical structures to optimize morphology may be undertaken.

This work may be extended to other synthesis techniques such as chemical vapor deposition or sol-gel synthesis among others, or compare Ni-Co hydroxide electrodes with other commonly used material for charge storage electrodes, such as manganese oxide or lithium-based electrodes. Extending LCA studies to other early-stage electrode material development could lead to environmentally-informed decisions in future research for energy storage.

### **3.5. Conclusions**

The present study identifies the main environmental hotspots in the synthesis of NCED-rGO electrode and its environmental performance relative to baseline NCED electrode and benchmark NCCP electrode. The analysis aims to support further design and optimization of this electrode

while giving a broader perspective on potentials of using rGO, and offering insight how this electrode performs in reference to current alternatives. The cradle-to-gate analysis was based on two functional parameters denoting capacity and lifetime, which are examined at different operational parameters of applied currents and criteria for end-of-life.

The findings suggest that the most dominant impacts in the fabrication of NCED-rGO come from electricity and cleaning, followed by GO, steel substrate and use of cobalt and nickel nitrates. Use of GO has shown to be advantageously applied to nickel-cobalt hydroxide electrodes as improved functionality of NCED-rGO electrode over NCED considerably outweighs added impacts from GO use. NCED-rGO is competitive with benchmark NCCP under specific circumstances.

We argue that the findings of this study need to be interpreted in view of the emerging nature of these technologies. The relative contribution of the impacts in the design of the electrode needs to be interpreted in view of changing potentials for scaling-up of reagent materials while comparative results need to be interpreted in view of technology readiness level of comparing electrodes and functional requirement of energy storage applications. The study recommends several resource strategies for further optimization of NCED-rGO.

## Chapter 4: Life cycle assessment of organic photovoltaic solar charger: a role of use intensity and irradiation <sup>3</sup>

### Abstract

Solar chargers for mobile phones are the first integration of organic photovoltaic (OPV) technology into commercial products. Although environmental impacts of OPVs have been studied extensively, the performance of chargers have been narrowly examined in reference to intensity of their use and use geographies. To explore these aspects, we study the environmental impacts of OPV chargers considering the charger as a substitute for a local electricity grid supply for charging a mobile phone. A consequential life-cycle assessment (LCA) was carried out to evaluate the environmental performance of the OPV charger in six European countries representative of different electricity grids and solar irradiation contexts. Particular effort is made to explore the implications of use intensity of the charger and determine a frequency at which charger is competitive. The results suggest that using an OPV charger has the potential to be environmentally friendly only in countries with high fossil-fuel share in their electricity supplies. The OPV charger is environmentally beneficial in Greece and Spain across most of the evaluated impact categories if used 100-120 times per year, which is practical given the high solar insolation in the two countries. Charging a phone with OPV in Germany or the Netherlands is environmentally-friendly only under conditions of intensive use of the device, or for selective impact categories. In the category of climate change, charging with OPV would represent an improvement in Greece and Germany. In two countries a phone-charging supported by OPV generates 2.5kg of CO<sub>2</sub>-equivalents per year in comparison to 2.9-3kg CO<sub>2</sub>-equivalents charging from the grid. Phone-charging supported by OPV in Norway and France is more impactful than using the grid for the majority of impact categories, including the category of climate change. The study contributes a novel methodology for looking at photovoltaic technology and helps inform users and policymakers who should consider the local context before an adoption of environmental technologies.

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<sup>3</sup> A version of this chapter was accepted for publication in the Journal of Cleaner Production as: Edis Glogic, Steffi Weyand, Michael P. Tsang, Steven B. Young, Liselotte Schebek, Guido Sonnemann (2019). Life cycle assessment of organic photovoltaic solar charger: a role of use intensity and irradiation. Journal of Cleaner Production, vol. 233, p. 1088-1096.

## 4.1. Introduction

Photovoltaic (PV) technology has been proposed as a more sustainable alternative to contemporary fossil fuel-based energy supply. Even though impacts are created during the manufacturing and disposal of PV products, overall improvements, especially in terms of greenhouse gasses mitigation, are significant (Serrano-Luján et al. 2017). From a range of photovoltaic technologies developed over several decades, the third generation organic PV (OPV) technology is advocated for superior eco-efficiency performance and distinct physical and electrochemical properties that could increase the range of PV products (Darling and You 2013; Hoppe and Sariciftci 2004). Compared to conventional silicon solar cells, OPVs have shown to have lower environmental impacts and shorter energy payback times (Espinosa et al. 2012; Espinosa, Garcia-Valverde, et al. 2011; García-Valverde, Cherni, and Urbina 2010; Roes et al. 2009; Serrano-Luján et al. 2017; Tsang, Sonnemann, and Bassani 2015; Yue et al. 2012), and when applied in the chargers for mobile phones (Alves dos Reis Benatto, Espinosa, and Krebs 2017; Tsang, Sonnemann, and Bassani 2016), portable lighting systems (Espinosa, García-Valverde, and Krebs 2011), and solar panels (Serrano-Luján et al. 2017; Tsang et al. 2016).

In practice, however, photovoltaics more often compete with other energy supply systems, in which case an aspect of the intensity of their use becomes more prominent and sometimes critical to their performance. Environmental impacts associated with the unit of PV electricity are created mostly during the production of PV device, while the use of PV devices when electricity is generated is virtually emission-free. Such disposition of impacts across life cycle phases of PV products prompts impacts to be lower with the more intensive use of PV device. Main factors influencing the use intensity of PV produces, are the choice of PV product integration and the geographical context of their application (i.e., solar irradiation).

The aspect of use intensity on perceived greenness of PV electricity supply presents a challenge to prospective product integration of OPV technology as a portable solar charger for the mobile phones. Even though these chargers integrate potentially greener OPV technology, they are used for only selective appliances such as mobile phones, headphones, cameras or other small electronic devices to facilitate on-demand charging in which instance the use could be expected to occur at a lower and intermittent frequency in comparison to stationary outdoors PV systems. OPV chargers are lightweight and portable and could easily be carried on person as a possible alternative to a powerbank charger and standard outlet supplying electricity from the local grid. Two studies that explored environmental impacts of using a charger, narrowly explore an aspect of charger use intensity and reach different conclusions. A study by Tsang et al. (2016) explored impacts of the charger in comparison to amorphous silicon as a substitute in which OPV was compared more favorably (Tsang et al. 2016). Benatto et al. (2017) investigated the OPV charger

as a substitute to a local electricity grid and amorphous silicon charger and has shown that OPV charger is not preferred to charge a phone in China and Denmark (Alves dos Reis Benatto et al. 2017). The results apply to the limited geographical scope and are based on a single use-intensity, largely neglecting intermittent use-profile of the charger, which is of particular concern to the results of the latter study where OPV is compared with electricity grid as a very different energy supply system. The competitiveness of OPV charger over amorphous silicon alternative was also ruled differently, which comes likely as a consequence of different assumptions of cell infrastructure, and expectations of efficiencies and lifetimes of the OPV cell.

Not conclusive to the studies on OPV chargers, modeling of intermittently used PV devices that resemble similar use behavior to that of PV chargers such as solar tents and solar backpacks, have not been performed to our knowledge. In the literature, intermittency of PV systems has been more readily discussed as a constraint to reliable energy supply (Margeta and Glasnovic 2012), and intermittency of solar irradiation (Laleman, Albrecht, and Dewulf 2013), rather than as a consequence of use-profile of PV device.

Taking aforementioned limitations, including the diverging results, geographical coverage and narrow use intensity assumptions of current studies on OPV charger, and also general lack of studies exploring intermittency of PV product use in assessment of environmental impacts, we investigate if the use of OPV charger as a substitute to the conventional electricity supply grid could reduce the impacts of charging a mobile phone. We look more closely at the device use intensity while exploring the broad geographic scope of Europe. The information is presented in the manner to achieve more comprehensive understanding of potential implications of the charger use while offering an original methodology to quantify the influence of solar irradiation for intermittently used PV devices. The methods and findings provided throughout this study could serve as valuable information to technology developers and policymakers who should consider product integration of this technology and the geographical context of its application.

## **4.2. Materials and methods**

The comparison between OPV charger and grid was carried out using consequential approach in LCA. Both direct and indirect environmental impacts considered through this approach are best suited for more perspective and context relevant assessment of emerging technologies and energy supply systems (Andersen 2013; Earles and Halog 2011; Liang et al. 2013).

Consistent with recommendations outlined in ISO 14040:2006 and ISO14044:2006 standards, LCA is carried out through, four phases: (1) goal and scope, (2) life cycle inventory, (3) life cycle impact assessment and (4) interpretation (ISO-14040 2006; ISO-14044 2006). The first two phases

are described in the current materials and methods section, and the third and fourth phases constitute the results section of this paper.

### *4.2.1. Goal and scope*

#### *4.2.1.1. Goal definition*

The goal of this study was to investigate the environmental consequences of using an OPV solar charger as a substitute for the electricity grid to charge a mobile phone in Europe, while specifically investigating the aspect of charger use-intensity and influence of irradiation on anticipated intermittent use. The study findings are expected to support OPV technology development and product integration.

#### *4.2.1.2. Functional unit*

The function for comparison between the OPV charger and the grid, is to charge a phone battery of 2000 mAh every day for five years. The selected capacity of 2000 mAh can be viewed either as charging a smaller battery or only partially charging a battery of bigger capacity. We consider this as a meaningful usage capacity considering the current designs of smartphones. As a reference, the iPhone 8 has a battery capacity of 1821 mAh, and the Samsung Galaxy S8 3000 mAh. The functional unit is one charged 2000 mAh battery using a standard 5V USB port. The reference flow is 10 Wh of electricity drawn and stored in the mobile phone battery.

#### *4.2.1.3. System boundaries*

The environmental analysis of the OPV charger device considers impacts arising from all life cycle stages including raw material extraction, manufacturing, use, and disposal. Assumptions of charger design and operating performances are adopted from previous works (Tsang et al. 2015, 2016). Included is a stand-alone 10 Wp (Watt-peak) solar charger (without battery power bank), with 0.2 m<sup>2</sup> of OPV panel and plastic casing. Additionally, this study includes a USB port which was not considered in a previous works due to a lack of data (Tsang et al. 2016). Consistent with Tsang et al. (2016), the structure of the OPV cell consists of two electrodes, an electron hole transport layer, an active layer, and a substrate. The active layer consists of fullerene derivative phenyl-C61-butyric acid methyl ester (PCBM) as a donor, and co-polymer polythiophene polymer poly(3-hexylthiophene) (P3HT) as an acceptor material, embedded in the form of bulk-heterojunction. Charge separation is facilitated using a transparent positive electrode of indium tin oxide, and the hole transport layer from molybdenum trioxide. A back electrode is from aluminum covered by the thin layer of lithium fluoride. A laminate is assumed from polyethylene terephthalate (PET). The OPV cell operates at 5% efficiency and five years lifetime, taken as a compromise between practical and laboratory performances (Tsang et al. 2015). Disposal of the charger was modeled assuming incineration, an established waste disposal route and dominant

waste treatment method for municipal solid waste in several countries in northern and western Europe (Blumenthal 2011). Incineration is only marginally better than landfilling a solar charger, another likely waste disposal alternative for the charger (Tsang et al. 2016). The charger is assumed to be used only for charging a mobile phone, and not the other electronic devices such as cameras or headphones.

#### 4.2.1.4. Impact assessment and interpretation methodology

The relative comparison between the OPV charger and the grid was carried out including (1) direct comparison and (2) break-even comparison. Moreover, the results from the two comparisons are interpreted in view of solar irradiation constraints. Comparison and interpretation approaches are represented by framework in Figure 4-1.

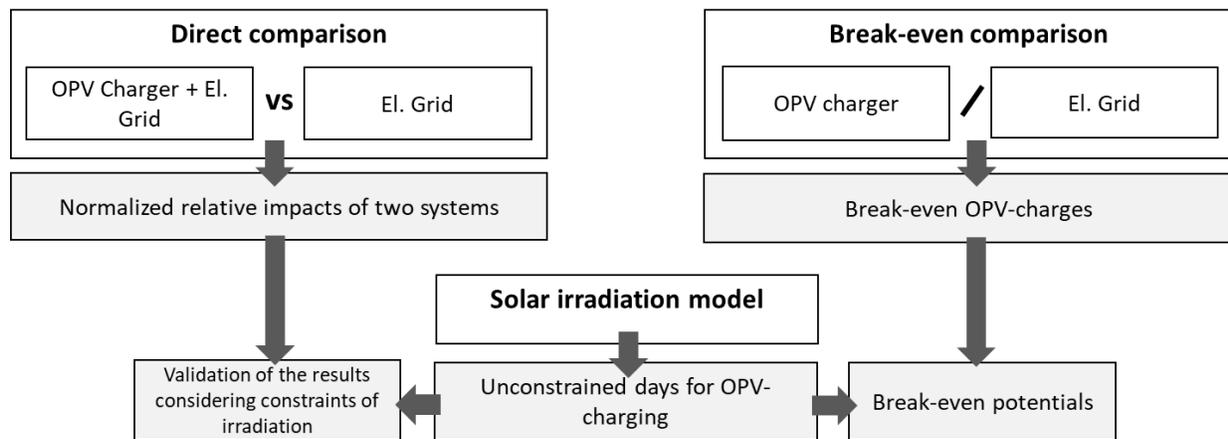


Figure 4-1. A framework describing comparative steps in this study and the irradiation model used for interpretation.

The direct comparison represents the conventional approach in LCA to calculate impacts between competing product systems using normalized values in the range 0-100. In this case, two product systems for charging a mobile phone are compared: (1) combining a solar charger and electricity grid, and (2) charging solely using the electricity grid. Charging with the solar charger is modeled at 150 times per year, the assumed use frequency adopted from the previous study on OPV chargers (Alves dos Reis Benatto et al. 2017). Over five years each product system supplies a total of 18.25 kWh of electricity, of which 7.5 kWh is drawn from the charger.

The break-even comparison, specifically developed in this study, describes the relative environmental impacts of the charger in reference to charger use intensity. Break-even comparison is designed to calculate phone charging frequency using an OPV charger, at which phone charging with the OPV charger (OPV-charges) would equal the impacts of charging with the grid (grid-charges). The break-even OPV-charges are calculated for each impact category using the following equation:

$$\text{break even OPV-charges} = \frac{\text{env. impact of production and disposal of OPV charger}}{\text{env. impact of single grid charge} \cdot \text{lifetime of OPV charger}}$$

The calculation of OPV-charges allows greater insight in the aspect of use intensity of the charger on its environmental performance and avoids making an assumption of charger use frequency as this is made in the direct comparison. Calculation of break-even OPV-charges could be established due to a different distribution of the environmental impacts across life cycle stages of the charger and grids. In the life cycle of the solar charger, all environmental impacts arise in the production and disposal phase, whereas most of the impacts of grid electricity are generated in the use phase (i.e., when fossil fuels are burned). A frequency of the charger use that exceeds break-even value would render the charger as more eco-efficient.

Interpretation of comparative results from the direct comparison and OPV charges is made through the lens of solar irradiation, given the sunlight as a limiting factor for charger use. We propose a method to incorporate solar irradiation constraints by calculating the number of *unconstrained days* per year which receive sufficient irradiation to fully charge a phone using a solar charger. *Nominal daily irradiation*, above which the day is unconstrained, represents solar irradiation sufficient to charge a 2000 mAh mobile phone battery using 10Wp OPV charger taking practical conditions such as technically required irradiation to charge a battery of given size, and also a portion of energy that wouldn't be utilized in practice. The extent of such unexploited energy would vary depending of irradiation strength and consistency, time of the day, and other practical factors that would obstruct the user from using a charger even when irradiation is available. Ideally, the value of nominal daily irradiation would also benefit from studies on user behavior to better understand how these practical constraints affect charging consistency, but in their absence in scientific literature, that value is assumed. The nominal daily irradiation is proposed as 2.5 kWh/m<sup>2</sup> of irradiation per day which equals to 3-4h of direct sunlight depending on the country and season and is 2.5 times greater than the theoretical irradiation needed to charge a phone battery<sup>4</sup>.

Using unconstrained days, it was possible to determine: (1) if OPV-charges set in direct comparison are appropriate, which is the case if an assumed value is lower than the number of unconstrained days for given country, and (2) the *break-even potentials* to express the likelihood of reaching break-even OPV-charges. Break-even potentials are calculated using the following equation:

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<sup>4</sup>The value of 1kWh/m<sup>2</sup> is derived by considering technical aspects of the charger and amount of energy needed to charge 2000mAh battery. Needed 10Wh of electricity is generated using 10Wp (peak) solar charger with panel area of 0.2m<sup>2</sup> operating at 5% efficiency: 10Wh/(1kWh/m<sup>2</sup>·0.05·0.2m<sup>2</sup>) =1kWh/m<sup>2</sup>.

$$\text{break-even potential} = \frac{\text{unconstrained days} - \text{break-even OPV-charges}}{\text{unconstrained days}}$$

According to the equation, the break-even potential has a value of zero if a number of unconstrained days are equal or lower than OPV-charges. The potential has a value of one if the number of unconstrained days is twice the number of break-even OPV-charges or greater.

Daily solar irradiation values, used to calculate unconstrained days, were extrapolated from monthly values of Global Horizontal Irradiance derived from the IRENA Global Atlas geographical coordinate grids and several measurement points for each of the six investigated countries (see Supporting Information (SI), Table S4-2). This irradiation value is expressed in Wh/m<sup>2</sup> and represents the total amount of solar irradiation received on the surface including both direct normal and diffuse horizontal irradiance. The daily irradiation values were extrapolated assuming a linear increase or decrease of irradiation throughout the month.

Emissions arising in the life cycles of the OPV charger and electricity grids were characterized using the ReCiPe 2008 Midpoint (H) (v1.11) impact assessment method, Table 4-1. The use of the method is in line with previous studies on OPV (Tsang et al. 2015, 2016), and an identified need for a broader set of categories and indicators in the modeling of PV and OPV systems (Laleman et al. 2013; Tsang et al. 2015). The comprehensive selection of impact categories included in the method was also needed to cover diverse range of impact-profiles characteristic for electricity grids in Europe. OpenLCA 1.6.3 open source LCA software was employed.

Table 4-1. Environmental impact categories of the ReCiPe midpoint method used in the study

Impact categories	Reference units	Abbreviations
Agricultural land occupation	m <sup>2</sup> *a	ALOP
Climate Change	kg CO <sub>2</sub> eq	GWP
Fossil depletion	kg oil eq	FDP
Freshwater ecotoxicity	kg 1,4-DB eq	FETP
Freshwater eutrophication	kg P eq	FEP
Human toxicity	kg 1,4-DB eq	HTTP
Ionizing radiation	kg U235 eq	IRP
Marine ecotoxicity	kg 1,4-DB eq	METP
Marine eutrophication	kg N eq	MEP
Metal depletion	kg Fe eq	MDP
Natural land transformation	m <sup>2</sup>	LTP
Ozone depletion	kg CFC-11 eq	ODP
Particulate matter formation	kg PM10 eq	PMFP
Photochemical oxidant formation	kg NMVOC	POFP
Terrestrial acidification	kg SO <sub>2</sub> eq	TAP
Terrestrial ecotoxicity	kg 1,4-DB eq	TETP
Urban land occupation	m <sup>2</sup> *a	ULOP
Water depletion	m <sup>3</sup>	WDP

### *4.2.2. Life cycle inventory*

Data on materials used in the manufacture of a 10 Wp OPV charger are taken from Tsang et al. (2015), and the inventory pertaining to incineration of the charger from Tsang et al (2016). All the assumptions for compilation of life cycle inventory is thoroughly described in the two studies, and are not repeated here. Only final values are disclosed in the supplement of this paper (Table S4-3) and materials used shortly described below. Data from the inventory, previously linked to the Ecoinvent v2.2 background data was linked to background data sourced from the Ecoinvent v3.3 consequential database for the average European context (Wernet et al. 2016).

Inventory of OPV charger assume production of PCBM via the pyrolysis technique using toluene as a feedstock. Deposition of all the layers in the OPV cell is assumed to be gravure printed, except for the transparent electrode that assumed the sputtering technique. Chlorobenzene is used as a solvent for the active layer application. Electricity is used for the annealing and printing of panel components and the lamination of the panel. The solar charger uses no produced energy or materials to operate and produces no direct emissions.

A dataset for a single USB port was obtained from Ecoinvent v3.3 as “market for electric connector, peripheral type buss -GLO”.

Data for the country-specific electricity grid mixes are from the Ecoinvent v3.3 consequential database for 2015 as “market for electricity, low voltage” (Itten, Frischknecht, and Stucki 2014; Wernet et al. 2016).

### *4.2.3. Selection of representative countries*

Charging scenarios were purposefully chosen to reflect on most diverse sources of electricity present in Europe with intention that broader conclusions can be made in regard to other regions in Europe and beyond. Two criteria were considered significant to the environmental performance of solar chargers: (a) greenhouse gas (GHG) intensity of the country’s electricity grid, and (b) annual solar irradiation available in the country.

GHG emission values were obtained from the Ecoinvent v3.3 consequential database (Itten et al. 2014; Wernet et al. 2016), and the yearly solar irradiation values were taken from the International Renewable Energy Agency’s Global Atlas (IRENA 2005) (see SI, Table S4-1, and Table S4-2).

Finally, out of 17 European countries for which both sets of data were available, six were selected (Figure 4-2) to represent each of the six partitions in the matrix of electricity supply grids and yearly solar irradiation. The electricity supply grid energy make-up of these countries is quite

variable with different single energy source having a high share in country's grid supply: Greece - 11% of oil, Spain - 26% of renewables, Germany - 44% of coal, the Netherlands - 42% of natural gas, France - 78% of nuclear and Norway - 96% of hydro. The GHG - irradiation performances of all 17 countries considered initially is disclosed in SI, Figure S4-1. Source data for Figure 4-1, Figure S4-1, and energy source share is derived from Table S4-1.

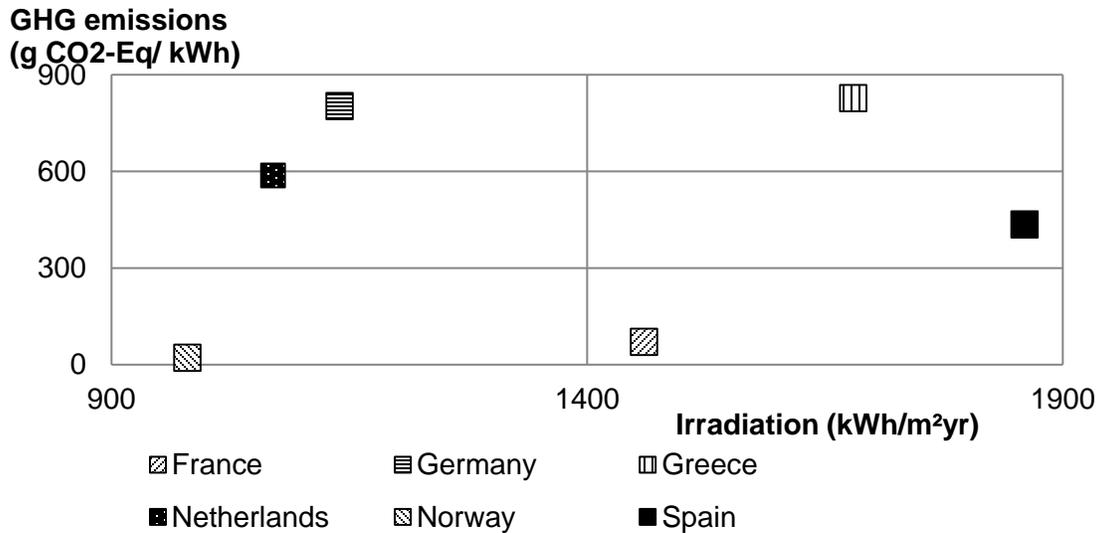


Figure 4-2. GHG-intensities of electricity supply grids and solar irradiation of six selected countries. Six countries cover a diverse range of possible charger use contexts, hence serve as a representative of Europe.

### 4.3. Results

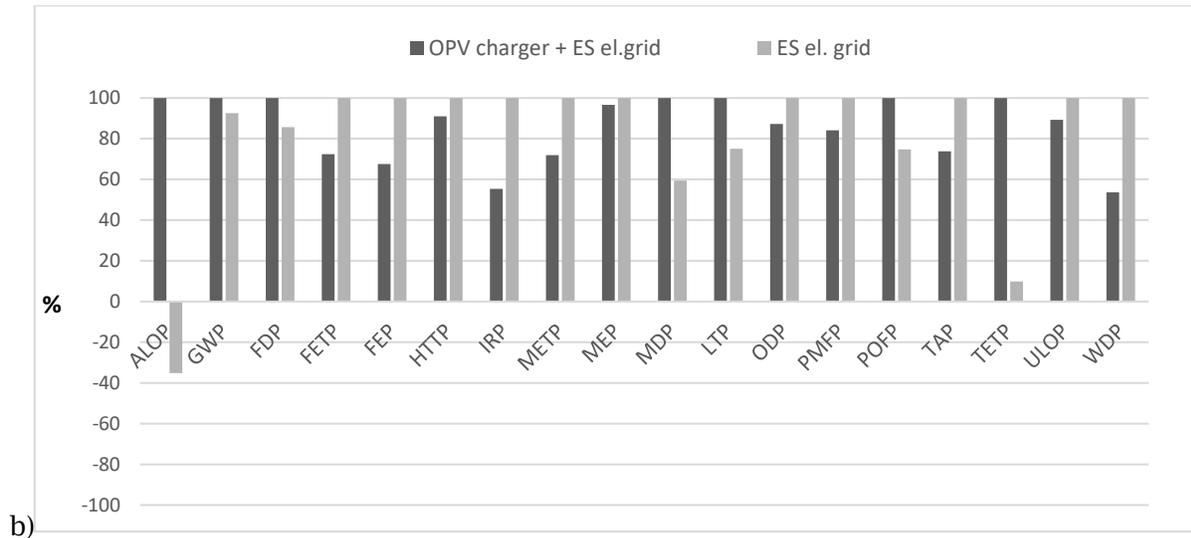
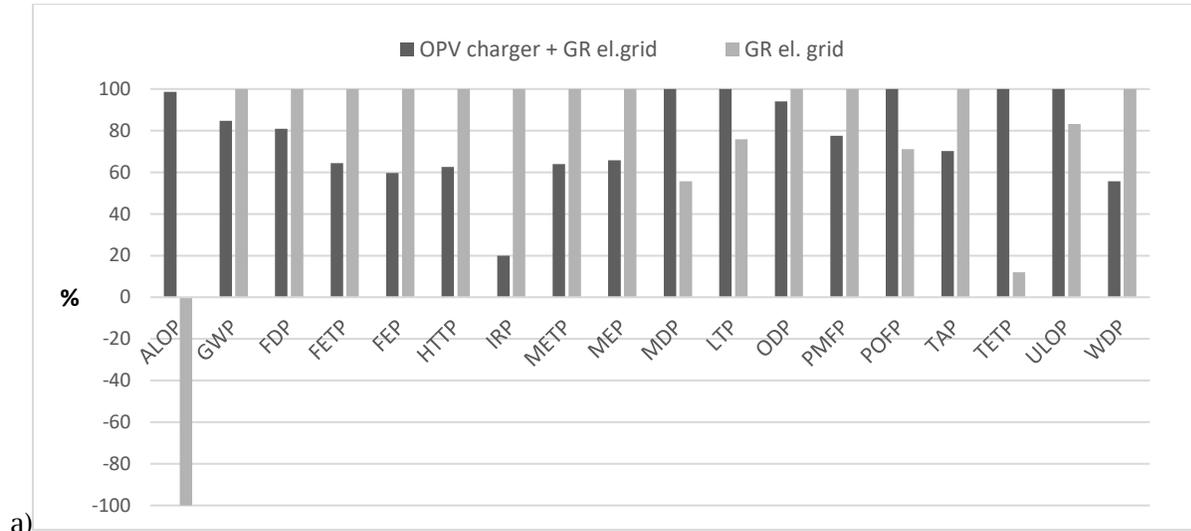
The results are presented in two sections. The life cycle impact assessment section, presents the findings from the direct and break-even comparison. In the interpretation section, findings from the direct comparison, and OPV charges determined through break-even comparison, are characterized for their validity and likelihood in view of solar irradiation capacity of investigated countries.

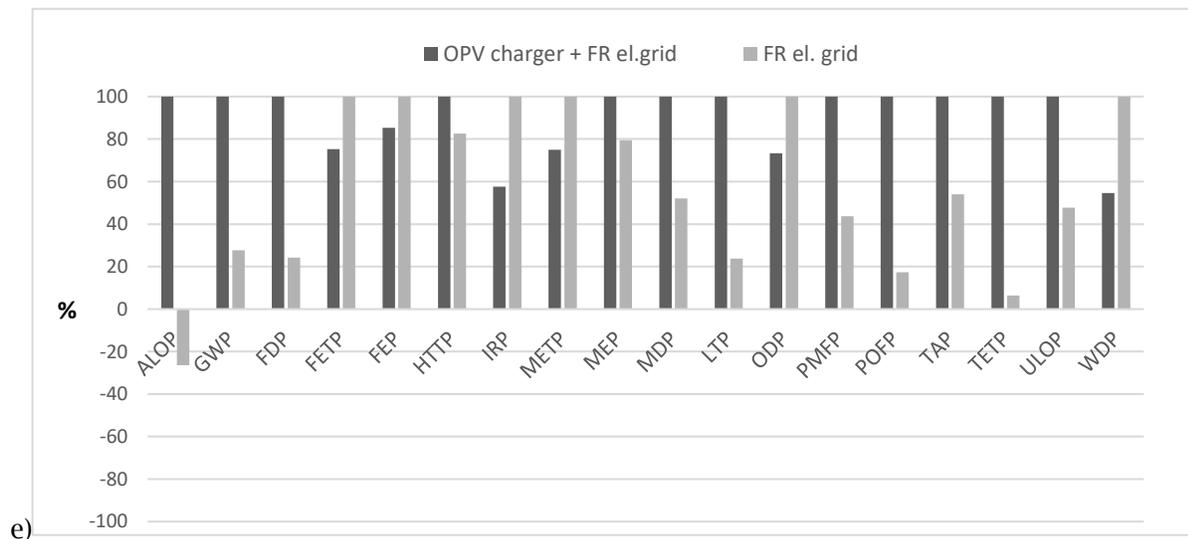
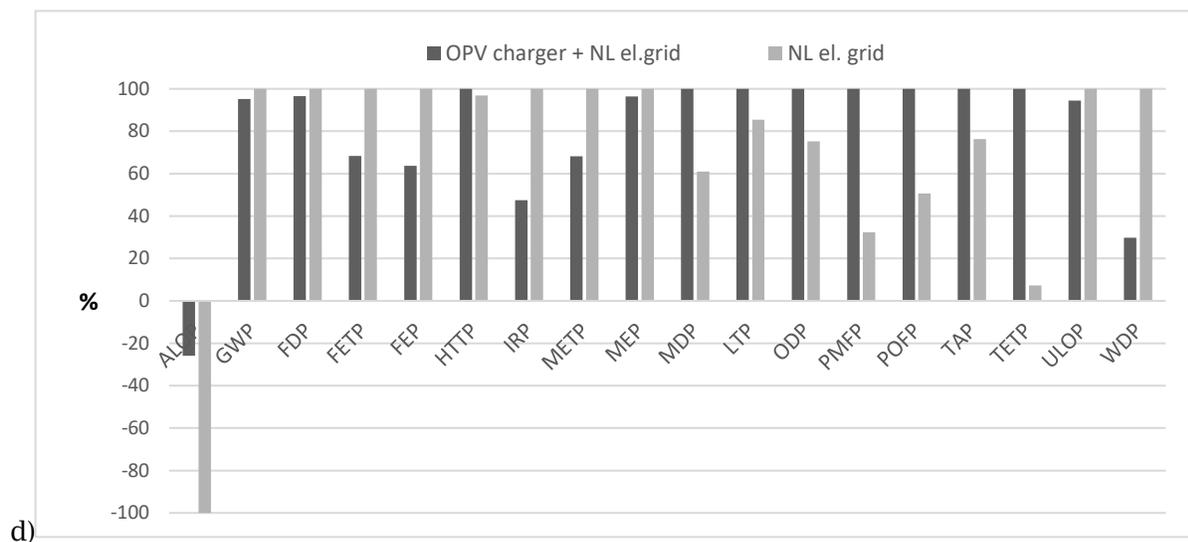
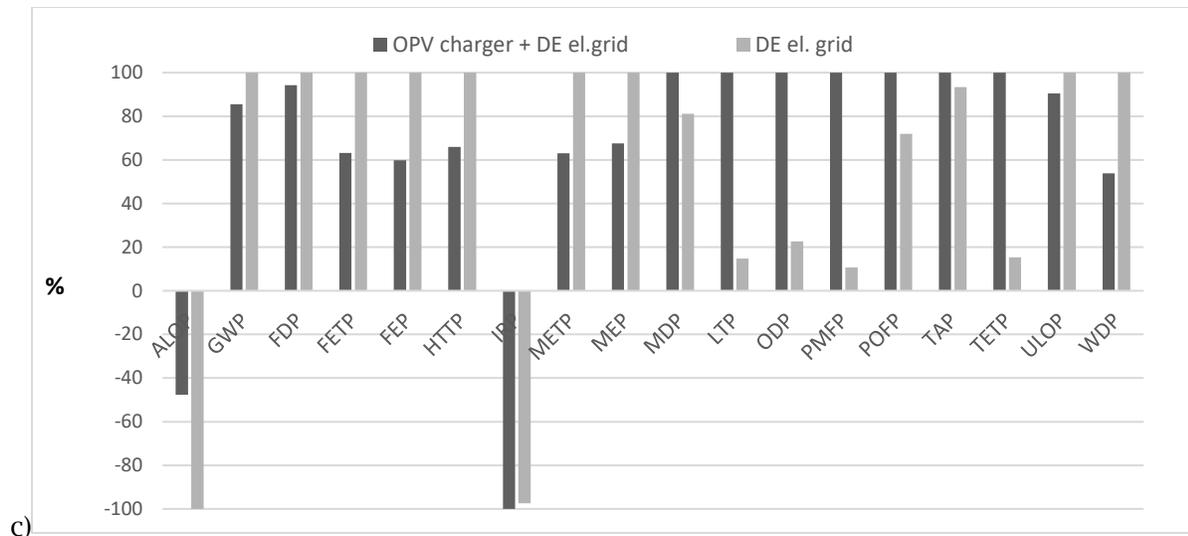
#### 4.3.1. Life cycle impact assessment

##### 4.3.1.1. Direct comparison

The relative comparison of a phone charged by combining OPV and grid electricity, versus grid-only charging is shown in Figure 4-3 (a-f) and absolute values are detailed in SI, Table S4-4, and

Table S4-5. Results show that the OPV-grid scenario appears competitive across most impact categories in Spain and Greece, and across eight of 18 categories in the Netherlands, ten in Germany, and six in France, while with only three categories showing benefits in Norway.





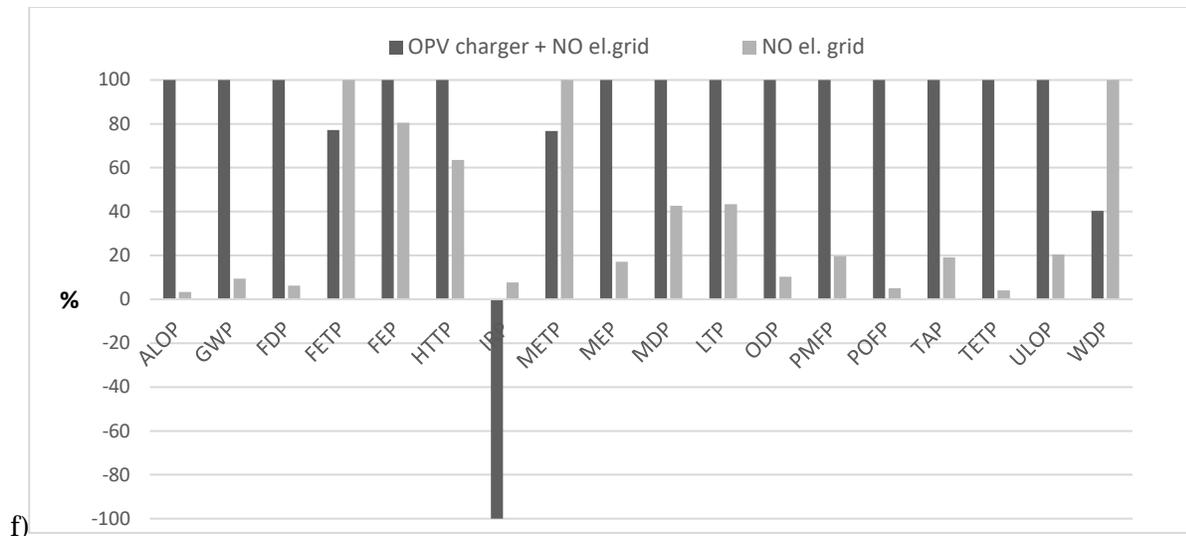


Figure 4-3 (a-f). Environmental impacts of charging of a 2000mAh phone battery every day for 5 years measured across 18 impact categories in six countries: a) Greece, b) Spain, c) Germany, d) the Netherlands, e) France, and f) Norway. Dark-colored bars show the results of combined OPV and grid-charging and lighter bars represent the grid-only system.

Use of OPV chargers is less beneficial in all countries across the potential category impacts of natural land transformation (LTP), ozone depletion (ODP), particulate matter formation (PMFP), and terrestrial ecotoxicity (TETP) due to impacts created as a result of polyester resin production for the charger casing. On the other hand, use of the charger lowers impacts across most of the water-related categories in all countries. That applies to the freshwater ecotoxicity (FETP), marine ecotoxicity (METP) and water depletion potential (WDP) for all countries, and marine and freshwater eutrophication impacts (MEP and FEP), for all countries except Norway. The environmental benefits in these categories are created from the avoided emissions of electricity due to OPV casing incineration. Potential impacts to categories of depletion of other resources provides mixed results. Metal depletion potential (MDP) is worse for the charger-use scenario in all countries, while fossil depletion (FDP) is similar for both product systems, except in France and Norway where electricity grids have notably lower impacts. A potential impact of low-voltage electricity grids in the category of agricultural land occupation (ALOP), comes with environmental benefits for all the countries due to the heat and power co-generation of biogas. Hence, those benefits are more pronounced in the grid-only scenario. The concentration of photochemical oxidants (POFP) that give rise to a summer smog is more impactful for the charger-grid scenario in almost all countries. For the particulate matter formation (PMFP) category, the charger-use scenario proves better only in Spain and Greece. Higher concentration of particulate matter in the electricity mix of both countries appears to be due to the use of lignite and coal. The OPV scenario is lower for ionizing radiation category (IR) due to energy recovery from the charger

incineration. Environmental benefits are also observed in the case of German electricity due to heat and power co-generation and the treatment of tailings in uranium milling. A use of OPV charger benefits the climate change (GWP) category in Germany, Greece and the Netherlands. Climate change (GWP), fossil depletion (FDP) and urban land occupation (ULOP) are similar for both grids and OPV charger and are likely to be sensitive to small deviations of OPV-charges above and below the 150 charges per year assumed for the comparison.

#### 4.3.1.2. Break-even comparison

Table 4-2 shows break-even OPV-charges. Below 100 OPV-charges the break-even points are reached in nearly all water-related impact categories in all countries except Norway, and in most of the impact categories for Greece. In Spain, breaks in most of the categories can be reached at around 100 OPV-charges. At around 130 OPV-charges roughly half of the impact categories could be reached for Germany and the Netherlands.

Table 4-2. OPV-charges to break-even with the environmental impacts of the electricity grids in six countries, across 18 impact categories.

<b>Impact category</b>	<b>GR</b>	<b>ES</b>	<b>DE</b>	<b>NL</b>	<b>FR</b>	<b>NO</b>
Agricultural land occupation	-	-	-	-	-	10527
Climate Change	94	179	97	133	1103	3640
Fossil depletion	80	211	129	137	1288	5592
Freshwater ecotoxicity	20	49	15	35	60	67
Freshwater eutrophication	3	32	3	18	96	238
Human toxicity	14	117	26	162	227	360
Ionizing radiation	0	0	160	0	0	0
Marine ecotoxicity	18	48	15	34	59	65
Marine eutrophication	25	138	32	137	244	1921
Metal depletion	439	400	235	384	487	641
Natural land transformation	266	271	2250	212	1319	627
Ozone depletion	128	103	1395	270	52	3345
Particulate matter formation	68	92	3204	912	622	1635
Photochemical oxidant formation	299	273	293	506	1899	7010
Terrestrial acidification	41	55	176	263	461	1691
Terrestrial ecotoxicity	2810	3453	2160	4756	5548	8910
Urban land occupation	223	111	115	130	550	1569
Water depletion	0	0	0	0	0	0

Break-even charges can only be derived for impact categories for which the more intensive use of solar charger leads to a reduction in the environmental impacts. Consequently, for impact categories where impacts of the grid charging are negative due to indirect environmental benefits,

the break-even values could not be implied. This is the case for the category of agricultural land occupation for all countries except Norway. Inversely, for impact categories where impacts of the OPV charging are negative due to environmental benefits, as such is the case for the categories of irradiation potential, and water depletion, impact categories are assigned zero value.

### 4.3.2. Interpretation of the results using solar irradiation constraints

#### 4.3.2.1. Characterization of OPV-charges used for the direct comparison

The unconstrained days were calculated as 305 in Spain, 282 Greece, 242 France, 205 Germany, 197 the Netherlands and 181 Norway. These values appear higher than the baseline assumption of 150 OPV-charges suggesting that the results shown in Figure 4-3 (a-f) are practical. However, given differences between assumed charges and unconstrained days in countries, results of the comparison for Spain, Greece and France are more conservative and thus more compelling than the conclusions derived for the Netherlands, Germany and Norway.

#### 4.3.2.2. Characterization of break-even OPV charges: break-even potentials

Break-even potentials are shown in Table 4-3. The high potentials (above 0.5) of achieving break-even OPV-charges applies to Spain and Greece, with the charger breaking even in majority of the impact categories. In the Netherlands and Germany, even though the break-even OPV charges can be achieved in most of the categories, the potentials of reaching break-even values are small. For example, for the Netherlands, in five of ten categories where OPV break-even charges could be achieved, the potentials are below 0.34. For Norway and France, most of the impact categories are not attainable. However, the break-even potentials in the remaining categories in France are high, suggesting a high likelihood of making improvements in specific categories by using the charger.

Break-even potentials mostly allow to observe relative likelihood among countries to reach break-even OPV-charges and highlight that similar break-even values have different potentials to be reached depending of country's irradiation. For instance, for Greece and Germany the break-even values of the category of climate change (94 and 97, respectively), although similar, translate in to higher potential for Greece (0.67) than Germany (0.53).

Table 4-3. Break-even potentials showing the relative likelihood of reaching OPV-charges.

	Break-even OPV-charging potentials					
	GR	ES	DE	NL	FR	NO
Agricultural land occupation	0.00	0.00	0.00	0.00	0.00	0.00
Climate Change	0.67	0.41	0.53	0.32	0.00	0.00
Fossil depletion	0.72	0.31	0.37	0.30	0.00	0.00

Freshwater ecotoxicity	0.93	0.84	0.93	0.82	0.75	0.63
Freshwater eutrophication	0.99	0.90	0.99	0.91	0.60	0.00
Human toxicity	0.95	0.62	0.87	0.18	0.06	0.00
Ionising radiation	1.00	1.00	0.22	1.00	1.00	1.00
Marine ecotoxicity	0.94	0.84	0.93	0.83	0.76	0.64
Marine eutrophication	0.91	0.55	0.84	0.30	0.00	0.00
Metal depletion	0.00	0.00	0.00	0.00	0.00	0.00
Natural land transformation	0.06	0.11	0.00	0.00	0.00	0.00
Ozone depletion	0.55	0.66	0.00	0.00	0.79	0.00
Particulate matter formation	0.76	0.70	0.00	0.00	0.00	0.00
Photochemical oxidant formation	0.00	0.10	0.00	0.00	0.00	0.00
Terrestrial acidification	0.85	0.82	0.14	0.00	0.00	0.00
Terrestrial ecotoxicity	0.00	0.00	0.00	0.00	0.00	0.00
Urban land occupation	0.21	0.64	0.44	0.34	0.00	0.00
Water depletion	1.00	1.00	1.00	1.00	1.00	1.00

Break-even potentials in range 0.5-1 signify high potentials, and 0-0.5 low-to-medium likelihood to reach OPV-charges. Potentials with the values of zero represent categories for which break-even value could not be achieved as break-even charges are greater than unconstrained days.

## 4.4. Discussion

Contrary to the previous studies (Espinosa et al. 2012; Espinosa, Garcia-Valverde, et al. 2011; García-Valverde et al. 2010; Roes et al. 2009; Serrano-Luján et al. 2017; Tsang et al. 2015; Yue et al. 2012), our research shows that OPV technology is not always environmentally-friendly and that the choice of integrating PV products plays a decisive role. In most of the investigated countries, the intensive use of charger is needed if charging with OPV is to be considered an improvement. Even in countries with dirtier grids, such as Greece where electricity grid supply is dominated by coal, and in Spain where grid supply is mostly based on use of oil, coal, and biomass, the charger needs to be used on average 100 times to have equal impacts with competing grids, and more intensively to be categorized as “green”. Overall, the OPV charger is more suited for targeting improvements in selective impact categories, rather than seeking to obtain improvements in all categories. Thus, given priority to specific impact categories, the charger could also be preferred in Germany, the Netherlands, and even France.

An observation to favorable charger performance for category of climate change in countries with dirtier electricity grids, echoes in earlier works where the charger was rated worse in Denmark, which has a high ratio of wind power, and positively in China where there is a high share of fossil fuels in the electricity grid (Alves dos Reis Benatto et al. 2017). However, for other impact categories our results vary which likely come about as a result of different assumptions for OPV cell design, lower operating efficiencies assumed, or the different version of Ecoinvent database

used for modeling (Steubing et al. 2016). The type of analysis that considers geographic variables for renewable energy is similar to work being undertaken to compare electric vehicles with cars with internal combustion engine (Nealer, Reichmuth, and Anair 2015). However, electric vehicles do better on cleaner grids, whereas OPV chargers compare better in the context of polluting grids. Principally, if CO<sub>2</sub> emission-equivalents are presumed as indicative of fuel share of electricity (refer to Figure 4-2 and Figure S4-1), our findings could be extended to assume charger performance in other countries with similar solar irradiation potentials and fuel shares of their grid supplies. In that case, the environmentally advantageous use of OPV charger within the reasonable frequencies of charger use could be achieved in Italy and Portugal. Use in the Czech Republic, the United Kingdom, and Luxemburg will result in environmental trade-offs between similar impact categories, whereas, the use of charger in Switzerland, Slovakia, Austria and Belgium would not be accommodating to low-impact phone charging using OPV.

The type of analysis we presented in our study is the first attempt to model the aspect of intermittency of PV devices as a feature of the product use-profile, the aspect which is highly uncertain and a more expected feature of emerging technologies, since a credible estimate of user behavior is more difficult. While the most conventional way to tackle this issue is to assess multiple assumption of charger use involving multiple scenarios and functional proxies, we offer an approach where the estimate of product use can be avoided altogether. Additionally, the demonstrated break-even comparison allows incorporating solar irradiation in the modeling of chargers. Lastly, this novel distance-to-target representation of the results generates information more palatable to the user, hence appealing to circular economy perspective where product user can take more proactive role. Similar approach to modeling could be applied to any consumer product whose performance changes with intensified use.

A main limitation of our work is associated with the assumption of nominal daily irradiation used to derive unconstrained days, that could not be well supported in the current literature on consumer behavior. Although, this is not detrimental to our overall findings as the preference across investigated impact categories is mostly divided between grids and an OPV charger, hence, small to medium variations in solar irradiation are expected to have minor influence on the results. Also, it is important to note that a technical durability of the charger (i.e., five years), although realistic assumption of technology (Green et al. 2017; Peters et al. 2011), is not necessarily an indication of the actual longevity of use (Khan et al. 2018). Both nominal daily irradiation value and expected lifetime assumptions could benefit from behavioral science and agent-based modeling that is increasingly used in environmental studies to estimate consumer behavior (Raihanian Mashhadi and Behdad 2018; Di Sorrentino, Woelbert, and Sala 2016). Another viable approach to realize potential for OPV-charging is with the help of ambient light sensors in mobile phones that can inform on user exposure to solar irradiation (Schuss et al. 2014).

Finally, when considering the prospective advantages between OPV chargers and the electricity grid, it is worth noting the differences between the two supply systems in terms of practical considerations like reliability and scale of energy provision. Solar chargers provide the convenience of outdoor charging, and in areas where charging is otherwise not accessible such as developing countries where grid infrastructure is not available. This flexibility and the potential of environmental performance in given countries would make portable OPV systems competitive replacements for diesel generators. On the other hand, grid electricity is often a more reliable electricity source that cannot be entirely replaced by a solar charger. The cost of electricity pertaining to both systems and the social aspects connected to resource use would need to complement this environmental analysis to fully support policy or consumer decision.

## 4.5. Conclusions

The study was carried out to determine if the use of OPV charger provides an improvement over conventional charging of the mobile phone in several countries in Europe while considering the frequency at which the charger is used. Comparison with conventional grid-charging is carried out both for an estimated use-rate of the charger, and inversely by calculating the use rate at eco-efficiency break-even points. Subsequently, the results from both comparative approaches are interpreted accounting for capacity of solar irradiation.

The findings suggest that OPV charger has the potential to be environmentally-friendly in the countries with dirtier electricity supplies and for targeting improvements in select impact categories. Overall, the use of OPV chargers could reduce impacts in water-related categories and increase impacts in categories representing atmospheric pollution. The OPV charger is beneficial in Spain and Greece but cannot compete with low-impact hydro and nuclear power of the grids in Norway and France.

The approach presented in this study constitutes a guiding framework for assessment of intermittently used products and offers a quantitative method for incorporating solar irradiation in modeling of PV products.

## Chapter 5 – Discussion and conclusions

This work helps fill the two research gaps identified in chapter 1: (i) the lack of experience and direction on combining CE indicators with LCA, and (ii) a general weakness in guidance available on applying LCA to emerging technologies. Thus, the research extends the scope of LCA in two directions: by considering broadening the scope of indicators to include indicators from the new economic paradigm of the Circular Economy (CE); and by providing new insights on using LCA for assessment of emerging technologies. The application of LCA in these two areas are identified in the scientific community (CIRAIG 2015; Haupt and Zschokke 2017; Kirchain Jr et al. 2017; Smith et al. 2019; UNEP 2011), and its potential further framed in this research as an effort for extending the scope of LCA as a sustainability assessment tool (Guinée 2016; Klopffer 2008; Sala et al. 2013a; Zamagni, Pesonen, and Swarr 2013).

The three case studies contributed to the objectives and the research questions outlined in Chapter 1. The first objective was met by investigating the capacity of the Material Circularity Indicator (MCI) to complement environmental assessment with LCA given potential trade-offs among MCI and LCA impact categories and indicators, applied to the case of alkaline batteries. The second objective was met by using LCA to evaluate emerging technologies for the cases of electrodes and OPV chargers. Assessment of three energy materials explored by three studies helps identify improvements to these specific important technologies.

This chapter gives an overview of the main contributions of this research, considers implications, discusses its limitations, and offers some recommendations for future research. Sections 5.1 and 5.2 provide a summary of the main findings discussed in view of the proposed objectives and research questions and discuss their implications to the broader literature theory and practice. Section 5.3 discusses the limitations of our research, and Section 5.4. provides recommendations for future research drawing on existing literature on CE indicators and emerging technology assessment, and the observations from the three case studies.

### **5.1. Challenges and opportunities for combining life cycle assessment with MCI**

Objective 1 was to evaluate the methodological potential of CE-indicators to complement environmental assessment with LCA. The two research questions under objective 1 were:

1-1. What are the challenges in combining LCA with circularity indicators, focusing on impact-circularity trade-offs and methodological differences?

1-2. What methodological improvements can be suggested to address these challenges?

To fulfil this first objective, the alkaline battery case study was used to examine the possibility of combining LCA results with a circular economy indicator. Several design and management scenarios for batteries were investigated with each scenario integrating a specific circularity strategy in comparison to the baseline so that the influence of specific strategies on the batteries' environmental and circularity performance could be differentiated. Strategies reviewed encompass two alternative recycling routes and several improvement strategies: use of recycled content in battery manufacture, increased used battery collection, and an improved battery lifetime (i.e., utility). In response to the research question 1-1, two challenges were noted: (i) trade-offs between MCI and LCA categories and indicators across two recycling routes and for several improvement strategies, and (ii) different sensitivities of MCI and LCA indicator values to particular modeling assumptions.

The trade-offs were largely observed for the recycled content use scenario, while improving utility, and battery collection in most cases resulted in improved circularity and reduced impacts, albeit to different degrees. Also, the trade-offs between LCA impact categories and indicators and MCI seems to be case- rather than strategy-specific (Lonca et al. 2018; Niero and Kalbar 2019).

In regards to the robustness of LCA impact categories and indicator values and MCI scores to a changing assumption for the system boundaries, we established that a decision to include certain byproducts of recycling differently affects two sets of values. Specifically, for recycled content scenarios, the assumption of a displacement of clinker cement with manganese slag has little effect on LCA results, but the MCI value changes in order of 30%. This disproportional sensitivity between LCA impact categories and indicators and MCI scores stems from the limitation of MCI to characterize material quality losses (EMF 2015; Saidani et al. 2017), whereas the characterization of material quality losses in LCA is indirectly accounted for through allocation of byproduct of recycling. Given the quality of byproducts, system is either credited for avoided emission for producing primary material (i.e., material moves in a closed-loop fashion), or for avoiding production of lesser-quality products, thus signaling if the recycling is “functional” or the material is downcycled. With the example of alkaline batteries, we now show the “gravity” of that particular disparity and how much that might affect the robustness of the results in such combined analysis. To improve the characterization of material quality losses in MCI, suggestions were made to consider use of complementary CE-indicators better suited to address material flows at end-of-life stage as they look more carefully in the nature of byproduct of a recycling. In that regard, following works are indicated as relevant: (Huysman et al. 2017; Linder et al. 2017;

Di Maio et al. 2017; Di Maio and Rem 2015; Park and Chertow 2014; Vanegas et al. 2018; Zink et al. 2016).

In response to identified challenges of combining LCA impact categories and indicators with MCI, as per research question 1-2, we propose a better visualization and interpretation of MCI values to improve understanding of trade-offs across strategies and comparison among categories and indicators (Figures 2-4, 2-5 and 2-6). MCI scores of different circularity strategies are normalized to their maximum circularity potential (i.e., a combined value of all identified applicable strategies over the life cycle of a product) to allow representation of MCI scores relative to what is practically achievable in terms of closing loops of resources. The proposed approach contributes to the development of the MCI indicator and provides an alternative to other efforts in comparing LCA impact categories and indicators with MCI (Lonca et al. 2018; Niero and Kalbar 2019; Walker et al. 2018). In these studies, MCI scores are normalized to their baseline scenario, while normalization in our method is based on the cumulative (net) circularity scenario and more leverage is given to incremental improvements in circularity. The new approach is arguably more suitable to compare circularity progress of different product assortments on an equal basis, as part of monitoring a company's internal progress in the adaptation of CE among products.

The implications to quality characterization of material losses, and the new approach for visualization, contribute to previously identified gap in the literature related to the lack of insight in CE indicators combining with LCA, as the means of indicators use and development (Lonca et al. 2018; Niero and Kalbar 2019; Walker et al. 2018). These observations and suggestions should influence how MCI is used and developed in the future and might be significant given that MCI is likely the most popular choice amongst micro/product-level CE indicators considering its early conception and origins, sophisticated construct, a whole life-cycle, and multiple criteria approach (Elia et al. 2017; EMF 2015; Moraga et al. 2019). On the other hand, this contribution is limited to qualification of indicator to measure circularity. Saidani et al. (2017) have shown that MCI lacks consistency with some of the CE principles including modularity, connectivity, upgradability, considerations for design and disassembly as a preventive maintenance of products and more granular levels of recovery such as remanufacturing or refurbishment (Saidani et al. 2017). Moraga et al. (2019) also note that none of CE-indicators, including MCI, support preservation of functions instead of products, and do not have a means to quantify product sharing, schemes for product redundancy, and multifunctionality (Moraga et al. 2019). However, the authors acknowledge MCI's applicability to industry, life cycle thinking approach, and sustainability perspective (Elia et al. 2017; Moraga et al. 2019; Saidani et al. 2017). MCI's future competitiveness and way of application will depend on how the next indicator developments respond to these critiques.

## 5.2. Application of life cycle assessment for emerging technology development

Objective 2 was to evaluate the use of LCA to improve the development of emerging energy materials. The two research questions under objective 2 were:

- 2-1. What are the environmental sustainability implications of new energy materials including opportunities/aspects for optimization across product life cycle, and when compared with conventional alternatives?
- 2-2. What are challenges and methodological approaches for improving assessment of emerging technologies?

This second objective was met through LCA case studies of two energy technologies: Ni-Co hydroxide charge storage electrodes, and organic photovoltaic (OPV) chargers. The case study of emerging Ni-Co hydroxide charge electrodes entails comparison between several synthesis methods to manufacture the electrodes to be used as an anode in supercapacitors or batteries. Synthesis is enabled by using electrodeposition with an optional use of graphene oxide. It was determined that environmental hotspots of electrodes are associated with electricity use, cleaning agents, and graphene oxide. The new synthesis method is less competitive with more mature coprecipitation technique under variety of possible uses although with small optimization of materials and efficiency improvement, the electrodes can be competitive given favorable operating parameters of current density and device lifetime expectations. The study of OPV chargers investigates the environmental impacts of chargers for replacing electricity grid for charging a phone in six countries in Europe. The chargers are potentially valuable substitutes to local electricity grids in three countries given frequent use and specific impact categories.

Each of two cases represents a valuable contribution given their implications for technology development and importance of renewable energy systems for sustainability (Glogic, Adán-Más, et al. 2019; Glogic, Weyand, et al. 2019). For example, understanding of process optimization hotspots for charge storage electrodes, new data inventory, and the outlook of (beneficial) use of graphene oxide in design of the electrodes, are valuable to the materials science research. Similarly, the insights into the deployment of OPV technology for the charger product-integration and investigated geographies, and the solar irradiation model for modeling of intermittently used PV devices, are valuable contributions to the field of environmental-impact modeling and future development of these technologies.

In the case of electrodes, as a very early-stage technology, the challenges to modeling associated with emerging-nature noted previously in Chapter 1, Section 1.3.4 (i.e., challenges *i - v*), includes the forecasting of data to scaled-up process materials, evaluation of the results of comparison and contribution and function, functional unit and reference flow definition. This latter challenge manifests in increased uncertainty over the type of a battery system and operating parameters at which the electrodes would be deployed. As these different parameters imply different degradation phenomena and reduction in capacity over the lifetime of the electrodes, different quantities of material inputs are required to deliver the functional equivalent of energy storage. In the case of OPV chargers, the modeling of use-phase associated with uncertain use-frequency at which chargers would be deployed, also imposed a challenge to the definition of function, functional unit and reference flow.

Commonality of noted challenge in two studies, prompted further investigation and is a topic of following discussion that have broader implications to assessment of emerging technologies in LCA. The challenge of defining the function, functional unit and reference flow in LCA could appear for both emerging and current technologies but it is deemed to be compounded for assessment of emerging technologies (Hetherington et al. 2014). Uncertainties to technology's operating conditions and use-frequency observed in two studies refer to what Cooper (2003) identifies as challenges in allocation of reference flow to functional unit. Related to this challenge are uncertainties in assumed performance and lifetime of the product, and system dependency (i.e., an ability to capture functional interactions with other product systems) (Cooper 2003). In our cases, this variability relates to the amount of electrodes required to produce the reference flow that depends of operating parameters at which electrodes are deployed and the assumed lifetime. For OPV chargers, variability relates to how intensively chargers are used. This challenge occurs also in one LCA studies reviewed earlier (Table 1-3, chapter 1) indicating that observation in our studies are not uncommon in assessment of emerging technologies. The analysis of epitaxial graphene oxide reports uncertainty related to final application of technology (Arvidsson and Molander 2017), the challenges analogous to the ones noted in the study of electrodes.

According to Cooper (2003), multiple potential sources of error can arise when the function, functional unit, and reference flow do not reflect on the reality of a product system (Cooper 2003). Besides the challenge to allocation of reference flow to functional unit noted above, errors can arise when 1) assigning functional unit to the multiple functions, 2) carrying out (functionally equivalent) comparisons for substantially different products where strict focus on functionality loses the view of product differences in which the function is delivered, and 3) when functions are non-quantifiable (Cooper 2003). Further challenges to functional unit definition relate more commonly to emerging technologies. Pourzahedi et al. (2018) identify challenges in the ability to

capture complete effects of the new technologies and compare them with conventional technologies. Challenges for new technologies may also arise if a functional unit is not able to grasp increased functional requirements and consumption trends of particular technology; in which case an increasing demand for performance can outcompete the improvements of new technologies (Deng and Williams 2011; Kim, Kara, and Hauschild 2017).

To address the challenge of allocation of reference flow to functional unit in case of electrodes, multiple scenarios are conducted to investigate a broader range of possible functional set-ups of potential current density and lifetime expectations at which electrodes could be deployed. For OPV chargers, to resolve use-frequency uncertainties a break-even analysis was employed that allowed to show where OPV chargers would represent an improvement given frequency of use while offering an insight if such frequency is feasible given solar irradiation constraints. This led to identifying several countries where use of OPV chargers would be plausible. While break-even approach is not necessary when PV devices are directly compared (e.g. Tsang, Sonnemann, and Bassani 2016), an uninformed assumption of use-frequency is not adequate when a comparison is made with other energy systems which PV's are more likely to substitute in practice (e.g., the electricity grid). The break-even approach is less widely used in LCA, but arguably more frequent for energy systems. For example, break-even analysis was used to calculate normative mileage for electric cars (Nealer et al. 2015), and energy payback time for photovoltaics (Tsang et al. 2015). However, it is argued here that this approach is particularly viable for emerging technologies with uncertain use-context. The break-even approach also avoids assuming that a new technology replaces an existing one on a one-for-one basis, a potentially inaccurate assumption as it ignores practical trends and aspects of human behavior that could be particularly uncertain for emerging technologies (Cooper and Gutowski 2018).

From the limited set of studies analyzed and reviewed in this research, it appears that challenges to function, functional unit and reference flow may be shifting from definition of functional unit (error 1 according to Cooper (2003)) towards challenges to allocation of reference flow. If such observations hold true, as we are going from incumbent to emerging technology assessment, modeling efforts might be shifting from life cycle impact assessment phase (to investigate multiple functionalities) to the interpretation phase (to test uncertainties related to the challenges of allocation of reference flow to functional unit).

This observation is potentially significant given the desire to better understand how assessment of emerging technologies can be credibly carried out, and pointing to the opportunities and challenges that are underrepresented in the literature (Pourzahedi et al. 2018; Smith et al. 2019). Application of LCA methodology in development of emerging technologies has a great potential to improve environmental performance of products (Boothroyd 1994; Tischner et al. 2000),

particularly in domain of energy production and storage. The choice of functional unit in particular has significant influence on transferability and comparability of the results (Gargiulo, Girardi, and Temporelli 2017), and presents the greatest cause of variability in assessment of some emerging systems (Hischier, Salieri, and Pini 2017).

### 5.3. Additional considerations

A main limitation of this research, to both use-contexts of LCA, is a small number of studies that could undermine or exaggerate the importance of some of the observations made. For use of LCA for emerging technology development this could affect the perception of ISO-based LCA to adequately deal with new emerging-nature challenges. Similarly, challenges of combining MCI with LCA categories and indicators are likely not conclusive to the aspects we identified. Other aspects related to differences in indicator constructs, and non-linearity inherent to MCI calculation may also pose challenges. Eventually, future development of CE indicators might re-design indicators to become more conducive or restricting to their potential use with LCA (Elia et al. 2017; Linder et al. 2017; Saidani et al. 2017).

The second limitation concerns the broadening assumption regarding the use of MCI to complement assessment with LCA without providing some more specific characterization to how MCI increases the aptitude of sustainability assessment and improve upon socio-economic pillar. Broadening as defined in work of CALCAS could be accomplished “by better incorporating sustainability aspects and linking to neighbouring models, to improve their significance” (Heijungs, Huppel, and Guinee 2009, p7). However, the link between the utility of closing-loops inherent to circularity indicators, and the social, economic, and environmental pillars has not been extensively investigated. According to Moraga et al. (2019), MCI only partially accounts for environmental, economic and social aspects. Integral to this discussion is also the question of how MCI, and CE indicators in general, relate to resource categories in LCA such as abiotic resource depletion (Steen 2006), or resource indicators of emerging importance such as indicators of resource criticality important to the resource access in the economy (Gemechu, Sonnemann, and Young 2017; Graedel et al. 2012). In comparison with these categories and indicators, the MCI has an absolute range of value, it changes in non-linear fashion, and focuses on material flows instead of products, and as a result influences the design and management of products differently. To improve and reach high value of MCI requires that material fractions are managed to extent that high material yield is recovered and that product design and management practices constantly strive to increase that yield towards fully closing the loops. This is particularly the case in consideration to increasing “utility” above an industry’s average, that is non-linear and changing variable dependent of the current industry best practices. Given the

multi-criteria perspective on closing the loops of materials that respond to many possible circularity strategies, MCI has potential to influence product design and management beyond one-dimensional eco-efficiency improvements targeted by conventional indicators in LCA. Given that focus on product eco-efficiency in pursuit of sustainable production and consumption is, at best, limited (Figge et al. 2017), considering MCI along other resources indicators in LCA could represent an added value.

As a third consideration, in the context of the two use-contexts explored, is that the current research does not try to fill in the “gap” between LCA application to emerging technology assessment and use with CE-indicators. Assessment of circularity was not considered for emerging technologies, and circularity indicators investigated on the basis of how they support emerging technology assessment.

Lastly, it is worthwhile pointing out that two general limitations or critiques of LCA itself are relevant, involving the technocratic view and limited outlook on necessity of production (Moltesen and Bjørn 2018). These concerns are particularly pervasive in the current research where the underlying context presumes necessary role of emerging technologies in a sustainable energy transition. LCA is a useful tool for the assessment of technologies and comparison of pre-determined alternatives, but it does not look into the necessity and importance of the services technology delivers. This applies to energy, as to any other commodity, and its use is unlikely to be addressed to the extent needed for sustainability by solely improving technological efficiency. An improvement of eco-efficiency of the service could lead to the so-called “rebound-effect”, where with increased availability and affordability of technology as a consequence of technological improvement, more of it is used. The common example within energy domain is the use of electricity for lighting that despite significant technological improvements has remained constant (Tsao and Waide 2010). To tackle the rates of resource consumption and pressing issues of climate change it is important that social, political and institutional restructuring takes place. Within, the role and reliance on technology to address current environmental problems and material provision will be more or less pronounced. Less dramatic reforms are proposed in a steady-state and a green economy where technology, and particularly renewable energy technology plays a key role (Daly 1973; UNEP 2011). A more radical redefinition of socio-economic model and less technocratic view is included in paradigm of de-growth (Daly 1974; Jackson 2009; UNEP 2011; Victor 2012). In a circular economy, seen as an “intermediate” target to sustainability, transitions involve innovation in new technologies, optimisation of existing systems, and technology indirectly affecting the system (i.e., information technology) (Potting et al. 2017). Depending on what role technology plays out in sustainable development, so will LCA given its utility to inform new and existing technologies.

## 5.4. Conclusions and recommendations for future work

Several recommendations are provided for future work.

First, future research could explore constraints and opportunities of use of CE-indicators with assessment methods and LCA in particular. This includes understanding methodological aspects and resolving potential trade-offs in their joint use. Likely, simpler, single-criterion CE indicators are more conducive to coupling but they are limited to a range of resource strategies (or life-cycle stages) to which they can be applied, while more complex index-based indicators such as MCI require more detail examination to identify these limitations and opportunities. It is particularly worthwhile considering how CE indicators complement these methods in terms of “broadening” and “deepening” for sustainability, particularly as their contribution to social, environmental and economic pillars of sustainability is still fairly unexplored (Linder et al. 2017; Moraga et al. 2019; Pauliuk 2018). It would be worthwhile considering if circularity indicators encompass some of the aspect of “deepening” LCA, another condition noted to improve range of sustainability contribution. It is noted that “the primary physical (or “environmental”) mechanisms within the system are deepened to include social and economic mechanisms” (Heijungs, Huppes, and Guinee 2009, p23).

The second recommendation, also drawing from stated limitations of this work, constitutes the exploration of CE-indicators for their use for emerging technology development, thus bridging the “gap” between broadening approaches explored in this research. To that aim, characterization of circularity indicators at earlier stages of technology readiness level could be an interesting area of future research. The potential of existing indicators to fulfil that role or consideration of new indicators can be explored. Saidani et al. (2017) mentions increasing development of “nano-level” circularity indicators focusing on circularity of components and materials, rather than products.

As the third recommendation, we call for a more careful and precise use of the terminology like “ex-ante”, “prospective”, and “anticipatory” in emerging technology LCAs, and better framing of these terms to encourage their adequate use. This recommendation is based on observations taken from the scoping review of LCA studies (Table 1-2), in which many studies do not identify with the new terminology, but also from our own experience. Neither the study on electrodes nor on OPV chargers assumed either of the three terms. While “ex-ante” is perhaps the most suitable description, it is not exclusively used for analysis of emerging technologies (Buyle et al. 2019). In the study of OPV chargers, priority was given to characterization of study as “consequential”, which was arguably more relevant to use to imply a study delimitation. In the study of electrodes,

“ex-ante” terminology was not suitable as a product benchmark was not incumbent technology as this is mandated under this categorization (Cucurachi et al. 2018). For assessment of electrodes, the choice of comparison with another technology (at the pilot, rather than mature technology) was more meaningful in particular case, and considering researcher’s interest. Better classification of emerging-type LCAs and subsequent use of the terms could allow for better organization of the current and future knowledge in this area through literature reviews of relevant LCA studies (Arvidsson et al. 2018; Cucurachi et al. 2018). Clear differentiation between emerging-type LCAs is also needed to prevent that term is used as originally introduced. For example, Tsang et al. (2018) refer to LCA as “anticipatory”, although the study does not include uncertainty and stakeholder analysis associated with the particular classification (Wender et al. 2014). While, definition among different classifications has been to a good degree laid out for “anticipatory” LCA (Wender et al. 2012), further classification could be made for “prospective” (Arvidsson et al. 2018), and “ex-ante” approaches (Cucurachi et al. 2018). An alternative to naming considerations is declaring the technology readiness level in LCA studies as another plausible way to organize studies that assess emerging technologies and their needs in a consistent manner (Gavankar, Suh, et al. 2015).

The last recommendation coming from this work is directed to materials scientists who are urged to consider opportunities at different technology maturity levels (from conception to application of technology) where specific environmental improvements can be made and LCA can be used to aid technology development process. At the very early stage of technology development, functional optimization can go in hand with optimization of process steps, where selection, and substitution of materials and processes can be considered. For example, pinpointing hotspot materials within the process can create an opportunity for less impactful material substitutes, while testing how reduced quantities of hotspot material affect device functionality can inform the most optimal setup of functionality and impact reduction. To identify these opportunities, an attributional approach in LCA is more appropriate as it tries to estimate impact of technology for its contribution to global impact reservoirs, hence giving an insight to how “good” is the technology. The next step in product design should be to consider how the technology is most suitably applied in various product integrations and what technology does it substitute. Good environmental performance is a relative aspect as any product uses materials and energy to be extracted, refined, used and disposed of. Similar to what we demonstrated in our case study on OPV chargers, such investigation is more appropriate to be carried out using consequential LCA approach, which tries to estimate impacts given the context of application, and considers the market mechanism and both direct and indirect impacts associated with the new technology (Ekvall and Weidema 2004).

As materials research evolves, particularly to address sustainability challenges of energy supply, a viable, comprehensive, and science-based assessment tools will be needed to guide their development and use in view of influence on the environment and benefits to the society, while ensuring that the positives outweigh the negatives and the best available choices are prioritized. A circular economy and a progress in development of new technologies alike, need to ensure that the assumptions of environmental improvements through more circular flows of resources, or new technologies that enable renewable energy use, are grounded in life cycle thinking and multi-criteria-based decisions. As it was discussed and demonstrated in this work, LCA is a viable tool to meet these challenges given that its capabilities are extended and opportunities for application seized. In a pursuit of sustainability knowledge, through interdisciplinary lens of materials sciences, circular economy, and LCA, we attempted to contribute to these efforts.

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## APPENDICES

### Annex 1: Supplementary Information - Life cycle assessment of emerging Ni-Co hydroxide charge storage electrodes: impact of graphene oxide and synthesis route

This document provides all the background information of life cycle assessment carried out to compare three electrodes based on nickel-cobalt hydroxides, abbreviated as NCED, NCED-rGO and NCCP. This supplementary document includes the derivation of stoichiometric representation of active electrode materials which are used as basis to establishing some of the inventory data, the inventory itself normalized per 1g of electrode and functional unit of the study, and data adaptations from other studies modeled as a foreground process. The document further outlines all the absolute values of life cycle impact assessment and choice of background data from the Ecoinvent database given for the parameters for the functional unit and alternative parameters considered in scenario analysis.

Table S3-1

Calculation of stoichiometric formulae for NCED and NCED-rGO, and NCCP electrode, which is used to establish use of nickel and cobalt nitrates

<p>Electrodeposition reaction (NCED and NCED-rGO)</p>	<p><math>\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O} \rightarrow \text{Ni}^{2+} + 6\text{H}_2\text{O} + 2\text{NO}_3^-</math>  <math>\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O} \rightarrow \text{Co}^{2+} + 6\text{H}_2\text{O} + 2\text{NO}_3^-</math>            (Nickel, cobalt and water probably in the complex form of <math>[\text{Ni}(\text{H}_2\text{O})_6]^{2+}/[\text{Co}(\text{H}_2\text{O})_6]^{2+}</math>)            When cathodic current is applied (input of electrons), at the surface of the conductive substrate:  <math>\text{NO}_3^- + 7\text{H}_2\text{O} + 8 e^- \rightarrow \text{NH}_4^+ + 10\text{OH}^-</math>            There are many possible mechanisms for this reaction, we consider this one found in (Ash, Paramguru, and Mishra 2010; Delmas, Faure, and Borthomieu 1992)  <math>2\text{Ni}^{2+} + 4\text{Co}^{2+} + 12\text{OH}^- + 2(\text{NO}_3^-)^{2-} + 3\text{H}_2\text{O} \rightarrow 6\text{Ni}_{0.33}\text{Co}_{0.66}(\text{OH})_2 \cdot (\text{CO}_3^{2-}, 2\text{NO}_3^-)_{0.33} \cdot (\text{H}_2\text{O})_{0.5}</math>            In this case, since there is an excess of nitrate ions instead of carbonates, this is the preferential anion that gets intercalated.</p>
<p>Coprecipitation reaction (NCCP)</p>	<p><math>\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O} \rightarrow \text{Ni}^{2+} + 6\text{H}_2\text{O} + 2\text{NO}_3^-</math>  <math>\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O} \rightarrow \text{Co}^{2+} + 6\text{H}_2\text{O} + 2\text{NO}_3^-</math>            (Nickel, cobalt and water probably in the complex form of <math>[\text{Ni}(\text{H}_2\text{O})_6]^{2+}/[\text{Co}(\text{H}_2\text{O})_6]^{2+}</math>)            Addition of NaOH induces 2 Simultaneous reactions:            (1) <math>2\text{Co}^{2+} + \text{H}_2\text{O}_2 \rightarrow 2\text{Co}^{3+} + 2\text{OH}^-</math>; redox reaction consisting of two semi reactions:            (1.1) <math>\text{H}_2\text{O}_2 + 2e^- \rightarrow 2\text{OH}^-</math> Reduction reaction</p>

	<p>(1.2) <math>\text{Co}^{2+} \rightarrow \text{Co}^{3+} + \text{e}^-</math></p> <p>(2) <math>2\text{Ni}^{2+} + 4\text{Co}^{3+} + 12\text{OH}^- + 2(\text{CO}_3^{2-}, 2\text{NO}_3^-) + 3\text{H}_2\text{O}</math>  <math>\rightarrow 6\text{Ni}_{0.33}\text{Co}_{0.66}(\text{OH})_2 \cdot (\text{CO}_3^{2-}, 2\text{NO}_3^-)_{0.33} \cdot (\text{H}_2\text{O})_{0.5}</math></p> <p>(3) <math>\text{Na}^+ + \text{NO}_3^- \rightarrow \text{Na}^+\text{NO}_3^-</math> in solution.</p> <p>Note that water and carbonates come from the solution in order maintain charge neutrality and occupy the remaining insterslab space. Carbonates anions come into solution from atmospheric <math>\text{CO}_2</math> with which a spontaneous exchange occurs (Ash et al. 2010; Delmas et al. 1992)</p>
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Table S3-2

Life cycle inventory quantities of NCED, NCED-rGO and NCCP, including material and energy inputs and waste outputs indicated per 1 g of active material (AM) and per functional unit (FU)

<i>Flows (original naming from Ecoinvent unless modeled as a foreground system)</i>	<i>Unit</i>	NCED		NCED-rGO		NCCP		<i>Data source</i>
		Amount per 1g AM	Amount per FU	Amount per 1g AM	Amount per FU	Amount per 1g AM	Amount per FU	
nickel nitrate hexahydrate	g	8.30E-01	3.16E-02	7.90E-01	8.50E-03	7.60E-01	6.94E-03	Modeled as foreground
cobalt nitrate hexahydrate	g	1.66E+00	6.33E-02	1.58E+00	1.70E-02	1.54E+00	1.41E-02	Modeled as foreground
graphene oxide	g	0.00E+00	0.00E+00	1.00E+00	1.08E-02	0.00E+00	0.00E+00	Modeled as foreground
water, ultrapure	g	1.15E+03	4.39E+01	1.20E+03	1.29E+01	2.45E+02	2.24E+00	Ecoinvent
ethanol, without water, in 99.7% solution state, from ethylene	g	4.89E+01	1.86E+00	4.89E+01	5.26E-01	2.46E+02	2.25E+00	Ecoinvent
sodium hydroxide, without water, in 50% solution state	g	0.00E+00	0.00E+00	0.00E+00	3.44E-02	4.96E-01	4.53E-03	Ecoinvent
hydrogen peroxide, without water, in 50% solution state	g	0.00E+00	0.00E+00	0.00E+00	4.01E+00	2.90E-01	2.65E-03	Ecoinvent
polytetrafluoroethylene	g	0.00E+00	0.00E+00	0.00E+00	4.29E-04	5.00E-02	4.56E-04	Modeled as foreground
carbon black	g	0.00E+00	0.00E+00	0.00E+00	7.38E-05	1.50E-01	1.37E-03	Ecoinvent
steel, chromium steel 18/8	g	3.20E+00	1.22E-01	3.20E+00	8.50E-03	3.20E+00	2.92E-02	Ecoinvent
electricity, medium voltage	Wh	3.33E+02	1.27E+01	3.72E+02	1.70E-02	1.47E+02	1.34E+00	Ecoinvent
transport, freight train	t/km	3.93E-02	1.50E-03	3.99E-02	1.08E-02	1.90E-01	1.73E-03	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t/km	1.32E-02	5.04E-04	6.86E-03	1.29E+01	6.34E-02	5.78E-04	Ecoinvent

Table S3-3 (a-d)

Processes modeled as foreground systems including: a) nickel nitrate hexahydrate, b) cobalt nitrate hexahydrate, c) graphene oxide, d) polytetrafluoroethylene

a)

Nickel nitrate hexahydrate	Estimated from Ullmann's Encyclopedia of Industrial Chemistry (Hoydonckx et al. 2007)		
<i>Flow</i>	<i>Unit</i>	<i>Amount per 1kg</i>	<i>Data source</i>
water, ultrapure	g	3.10E+02	Ecoinvent
nickel, 99.5%	g	2.02E+02	Ecoinvent
electricity, medium voltage	kJ	8.25E+02	Ecoinvent
nitric acid, without water, in 50% solution state	g	4.33E+02	Ecoinvent
transport, freight train	t*km	3.81E-01	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t*km	1.27E-01	Ecoinvent

b)

Cobalt nitrate hexahydrate	Estimated from Ullmann's Encyclopedia of Industrial Chemistry (Hoydonckx et al. 2007)		
<i>Flow</i>	<i>Unit</i>	<i>Amount per 1kg</i>	<i>Data source</i>
cobalt	g	2.02E+02	Ecoinvent
water, ultrapure	g	2.48E+02	Ecoinvent
nitric acid, without water, in 50% solution state	g	8.66E+02	Ecoinvent
transport, freight, lorry 16-32 metric ton, EURO6	t*km	2.14E-01	Ecoinvent
transport, freight train	t*km	6.41E-01	Ecoinvent

c)

Graphene oxide	Sourced from (Cossutta et al. 2017)		
<i>Flow</i>	<i>Unit</i>	<i>Amount per 1kg</i>	<i>Data source</i>
lime, hydrated, loose weight	g	2.28E+04	Ecoinvent
graphite, battery grade	g	7.12E+02	Ecoinvent
potassium permanganate	g	2.14E+03	Ecoinvent
hydrogen peroxide, without water, in 50% solution state	g	1.24E+03	Ecoinvent
electricity, medium voltage	Wh	2.78E+03	Ecoinvent
water, ultrapure	g	2.24E+05	Ecoinvent
sulfuric acid	g	3.02E+04	Ecoinvent

d)

Polytetrafluoroethylene	Sourced from (Jungbluth et al. 2012)		
<i>Flow</i>	<i>Unit</i>	<i>Amount per per 1 kg</i>	<i>Data source</i>
refinery sludge	kg	4.39E+00	Ecoinvent
chlorodifluoromethane	kg	1.81E+03	Ecoinvent
municipal solid waste	kg	1.22E+00	Ecoinvent
transport, freight, lorry 7.5-16 metric ton, EURO6	t*km	2.30E-01	Ecoinvent
heat, district or industrial, natural gas	MJ	3.75E+04	Ecoinvent
heat, district or industrial, other than natural gas	MJ	4.68E+03	Ecoinvent
chemical factory, organics	Item(s)	4.00E-07	Ecoinvent

Table S3-4

Functional parameters of capacity and cycling stability (number of charge-discharge cycles) at current density of 1 A·g<sup>-1</sup>, 4 A·g<sup>-1</sup> and 10 A·g<sup>-1</sup>, and capacity fade of 20 and 30%.

	NCED	NCED-rGO	NCCP
	<b>Capacity</b>		
<b>Current density (A/g)</b>	<i>mAh/g</i>	<i>mAh/g</i>	<i>mAh/g</i>
1	30	96	121
2	26	58	114
4	22	49	102
10	15	43	96
	<b>Cycling stability</b>		
<b>Capacity fade (%)</b>	<i>#cycles</i>	<i>#cycles</i>	<i>#cycles</i>
20	972	1676	1006
30	1804	2557	2048

Table S3-5

Absolute values of NCED, NCED-rGO, NCCP and scaled-up scenario for NCED-rGO involving efficient use of graphene oxide.

Impact categories & indicators	Reference unit	NCED	NCED-rGO	NCCP	NCED-rGO scaled-up
Marine ecotoxicity	kg 1,4-DB eq	2.20E-04	6.60E-05	4.37E-05	5.28E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	1.39E-06	4.50E-07	2.74E-07	2.14E-07
Freshwater ecotoxicity	kg 1,4-DB eq	2.20E-04	6.71E-05	4.42E-05	5.44E-05
Fossil depletion	kg oil eq	4.52E-03	1.44E-03	2.56E-03	1.09E-03

Human toxicity	kg 1,4-DB eq	6.01E-03	1.85E-03	1.23E-03	1.36E-03
Water depletion	m3	7.63E-02	2.42E-02	1.23E-02	1.96E-02
Climate Change	kg CO2 eq	1.19E-02	3.97E-03	4.03E-03	2.80E-03
Ionising radiation	kg U235 eq	3.28E-03	1.06E-03	4.20E-04	8.50E-04
Metal depletion	kg Fe eq	1.91E-03	1.18E-03	4.80E-04	4.90E-04
Cumulative energy demand	MJ	2.81E-01	8.94E-02	1.27E-01	6.96E-02

Table S3-6 (a-c)

Relative impacts of NCED-rGO in comparison with NCED and NCCP when considering different combinations of current densities including 1, 4 and 10 A/g, and criteria for capacity fade of 20% and 30%. Percentage value indicate relative difference in impact for each category. Scenario abbreviation refer to combination of current density (CD) and capacity fade (CF): S-0 - CD 1 A·g<sup>-1</sup>, CF 20% (baseline, depicted in Figure 4 and 5); S-1 - CD 4 A·g<sup>-1</sup>, CF 20%; S-2 - CD 10 A·g<sup>-1</sup>, CF 20%; S-3 - CD 1 A·g<sup>-1</sup>, CF30%; S-4 - CD 4 A·g<sup>-1</sup>, CF 30%; S-5 - CD 10 A·g<sup>-1</sup>, CF 30%.Impacts of NCED-rGO are lower for percentage values preceded by the minus sign and are higher for positive values.

a)

Impact categories	Reference unit	NCED				
		S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	3.00E-04	4.40E-04	1.40E-04	1.90E-04	2.80E-04
Terrestrial ecotoxicity	kg 1,4-DB eq	1.90E-06	2.79E-06	9.02E-07	1.23E-06	1.80E-06
Freshwater ecotoxicity	kg 1,4-DB eq	3.10E-04	4.50E-04	1.40E-04	2.00E-04	2.90E-04
Fossil depletion	kg oil eq	6.16E-03	9.05E-03	2.92E-03	3.99E-03	5.85E-03
Human toxicity	kg 1,4-DB eq	8.19E-03	1.20E-02	3.88E-03	5.30E-03	7.77E-03
Water depletion	m3	1.04E-01	1.53E-01	4.93E-02	6.73E-02	9.86E-02
Climate Change	kg CO2 eq	1.63E-02	2.39E-02	7.71E-03	1.05E-02	1.54E-02
Ionising radiation	kg U235 eq	4.48E-03	6.57E-03	2.12E-03	2.89E-03	4.25E-03
Metal depletion	kg Fe eq	2.60E-03	3.86E-03	1.23E-03	1.68E-03	2.47E-03
Cumulative energy demand	MJ	3.83E-01	5.62E-01	1.82E-01	2.48E-01	3.63E-01

b)

		NCED-rGO
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Impact categories	Reference unit	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	1.20E-04	1.30E-04	3.33E-05	6.53E-05	7.44E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	7.94E-07	9.05E-07	2.28E-07	4.45E-07	5.08E-07
Freshwater ecotoxicity	kg 1,4-DB eq	1.20E-04	1.30E-04	3.39E-05	6.64E-05	7.57E-05
Fossil depletion	kg oil eq	2.54E-03	2.90E-03	7.30E-04	1.42E-03	1.62E-03
Human toxicity	kg 1,4-DB eq	3.25E-03	3.71E-03	9.30E-04	1.83E-03	2.08E-03
Water depletion	m3	4.26E-02	4.86E-02	1.22E-02	2.39E-02	2.73E-02
Climate Change	kg CO2 eq	7.01E-03	7.98E-03	2.01E-03	3.93E-03	4.48E-03
Ionising radiation	kg U235 eq	1.87E-03	2.13E-03	5.40E-04	1.05E-03	1.20E-03
Metal depletion	kg Fe eq	2.07E-03	2.36E-03	5.90E-04	1.16E-03	1.33E-03
Cumulative energy demand	MJ	1.58E-01	1.80E-01	4.52E-02	8.84E-02	1.01E-01

c)

		NCCP				
Impact categories	Reference unit	S-1	S-2	S-3	S-4	S-5
Marine ecotoxicity	kg 1,4-DB eq	4.67E-05	4.96E-05	2.35E-05	2.88E-05	3.06E-05
Terrestrial ecotoxicity	kg 1,4-DB eq	2.92E-07	3.11E-07	1.51E-07	2.43E-07	2.59E-07
Freshwater ecotoxicity	kg 1,4-DB eq	4.72E-05	5.02E-05	2.38E-05	2.88E-05	3.06E-05
Fossil depletion	kg oil eq	2.73E-03	2.90E-03	1.50E-03	1.82E-03	1.93E-03
Human toxicity	kg 1,4-DB eq	1.31E-03	1.40E-03	6.50E-04	8.00E-04	8.50E-04
Water depletion	m3	1.31E-02	1.39E-02	6.99E-03	8.40E-03	8.92E-03
Climate Change	kg CO2 eq	4.30E-03	4.57E-03	2.34E-03	2.90E-03	3.09E-03
Ionising radiation	kg U235 eq	4.40E-04	4.70E-04	2.40E-04	3.00E-04	3.20E-04
Metal depletion	kg Fe eq	5.20E-04	5.50E-04	2.60E-04	3.10E-04	3.30E-04
Cumulative energy demand	MJ	1.36E-01	1.44E-01	7.44E-02	9.04E-02	9.61E-02

## Annex 2: Supplementary Information - Life cycle assessment of organic photovoltaic charger use in Europe: the role of product use intensity and irradiation

This supplementary document contains background data for: 1) selection of the representative European countries to be investigated for context of OPV charger use, 2) development of method to indicate unconstrained number of days, based on irradiation profiles of selected countries, and 3) absolute values related to LCIA analysis, and process selection in conversion of pre-existing data published in Tsang et al (2015 & 2016), from attributional Ecoinvent v2.2 to consequential v3.3 (Tsang et al. 2015, 2016).

Table S4-1

Greenhouse gas (GHG) emission, average yearly irradiation, and electricity mix composition for 23 European countries

European IEA member states	Average yearly irradiation <sup>[1]</sup> kWh/m <sup>2</sup> yr	GHG emissions of electricity mix <sup>[2]</sup> g CO <sub>2</sub> -eq	Electricity mix, Data from 2015 <sup>[3]</sup> C: Coal, O: Oil, G: Gas, N: Nuclear, RE: Renewables, H: Hydro, B: Biofuels, W: Wind, G: Geothermal, S: Solar
Austria	1210	111.00	C: 8%,O: 1.4%, G: 13%, N: 0%, RE: 77.5%, H: 60%, B: 8%, W: 8%, S: 1.5%,
Belgium	1060	149.00	C: 6%,O: 0.3%, G: 33%, N: 38%, RE: 22%, H: 0%, B: 10%, W: 8%, S: 4%,
Czech Republic	1170	609.00	C: 53%,O: 0%, G: 3%, N: 32%, RE: 12%, H: 2%, B: 6%, W: 1%, S: 3%,
Denmark	NaN	15.00	C: 25%,O: 1%, G: 6%, N: 0%, RE: 68%, H: 0%, B: 17%, W: 49%, S: 2%,
Estonia	NaN	No data	C: 82%,O: 1%, G: 1%, N: 0%, RE: 16%, H: 0%, B: 9%, W: 7%, S: 0%,
Finland	NaN	104.00	C: 8%,O: 0%, G: 8%, N: 34%, RE: 44%, H: 24%, B: 17%, W: 3%, S: 0%,
France	1460	71.00	C: 2%,O: 0%, G: 4%, N: 78%, RE: 16%, H: 10%, B: 1%, W: 4%, S: 1%,
Germany	1140	803.00	C: 44%,O: 1%, G: 10%, N: 14%, RE: 30%, H: 3%, B: 9%, W: 12%, S: 6%,
Greece	1680	827.00	C: 43%,O: 11%, G: 18%, N: 0%, RE: 30%, H: 12%, B: 1%, W: 9%, S: 8%,
Hungary	1250	458.00	C: 19%,O: 0%, G: 17%, N: 52%, RE: 12%, H: 1%, B: 8%, W: 2%, S: 1%,
Ireland	960	No data	C: 17%,O: 1%, G: 44%, N: 0%, RE: 28%, H: 3%, B: 2%, W: 23%, S: 0%,
Italy	1500	467.00	C: 16%, O: 5%, G: 39%, N: 0%, RE: 39%, H: 16%, B: 8%, W: 5%, Geo: 2%, S: 8%,
Luxembourg	1130	758.00	C: 0%,O: 0%, G: 63%, N: 0%, RE: 37%, H: 7%, B: 14%, W: 8%, S: 8%,
Netherlands	1070	587.00	C: 39%,O: 1%, G: 42%, N: 4%, RE: 14%, H: 0%, B: 6%, W: 7%, S: 1%,
Norway	980	21.00	C: 0%,O: 0%, G: 2%, N: 0%, RE: 98%, H: 96%, B: 0%, W: 2%, S: 0%,
Poland	1100	1.00	C: 81%,O: 1%, G: 4%, N: 0%, RE: 14%, H: 1%, B: 6%, W: 7%, S: 0%,
Portugal	1790	565.00	C: 29%,O: 3%, G: 21%, N: 0%, RE: 49%, H: 17%, B: 7%, W: 23%, S: 2%,
Slovakia	1200	33.00	C: 13%,O: 1%, G: 6%, N: 57%, RE: 23%, H: 15%, B: 6%, W: 0%, S: 2%,

Spain	1860	435.00	C: 20%,O: 6%, G: 19%, N: 21%, RE: 36%, H: 10%, B: 3%, W: 18%, S: 5%,
Sweden	NaN	23.00	C: 1%,O: 0%, G: 0%, N: 35%, RE: 64%, H: 47%, B: 7%, W: 10%, S: 0%,
Switzerland	1240	10.00	C: 0%,O: 0%, G: 1%, N: 35%, RE: 64%, H: 58%, B: 4%, W: 0%, S: 2%,
United Kingdom	1030	696.00	C: 23%,O: 1%, G: 30%, N: 21%, RE: 26%, H: 2%, B: 10%, W: 12%, S: 2%,

<sup>[1]</sup> Extracted from <https://irena.masdar.ac.ae/GIS/?map=529#> for capitals, for countries with different climate more than one city was chosen (Austria, Croatia, France, Germany, Italy, Poland, Portugal, Spain)

<sup>[2]</sup> Calculated via openLCA 1.6.3, Ecoinvent 3.3 consequ., LCIA methods 1.5.6, ReCiPE, Climate change, Process country-specific "market for electricity, low voltage"

<sup>[3]</sup> IEA website (<https://www.iea.org/countries/membercountries/>) country-specific key energy data

Table S4-2

Monthly irradiation of investigated countries (in kWh/m<sup>2</sup>)

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
<b>Greece</b>	63	84	139	173	183	225	222	207	139	105	67	57
<b>Spain</b>	83	98	155	195	225	233	248	211	170	110	69	64
<b>Germany</b>	49	94	159	241	296	311	300	282	206	138	62	45
<b>NL</b>	19	39	72	131	168	183	142	125	101	57	21	14
<b>France</b>	107	118	172	206	244	264	289	270	211	162	112	97
<b>Norway</b>	7	22	74	136	159	182	165	125	74	29	6	0

Table S4-3

Direct material and energy consumption for producing 10 Wp OPV charger based on Tsang et al (2015; 2016)

Materials	Quantity	Unit	OPV component
PET	273	g	Casing material
Polyester	273	g	Casing material
PET	26.6	g	Lamination
PET	14.8	g	Substrate
Fluorine-doped tin oxide solution	0.36	g	Transport electrode
Molybdenum oxide	0.048	g	Hole transport layer
PCBM	0.041	g	Active layer
P3HT	0.047	g	Active layer
Chlorobenzene	1.532	g	Solvent for active layer

Aluminium	0.191	g	Back electrode
Lithium Fluoride	0.012	mg	Back electrode
<b>Energy</b>	<b>Quantity</b>	<b>Unit</b>	<b>Process</b>
Electricity	1.026	MJ	Annealing
Electricity	0.512	MJ	Printing of panel
Electricity	0.017	MJ	Lamination of panel

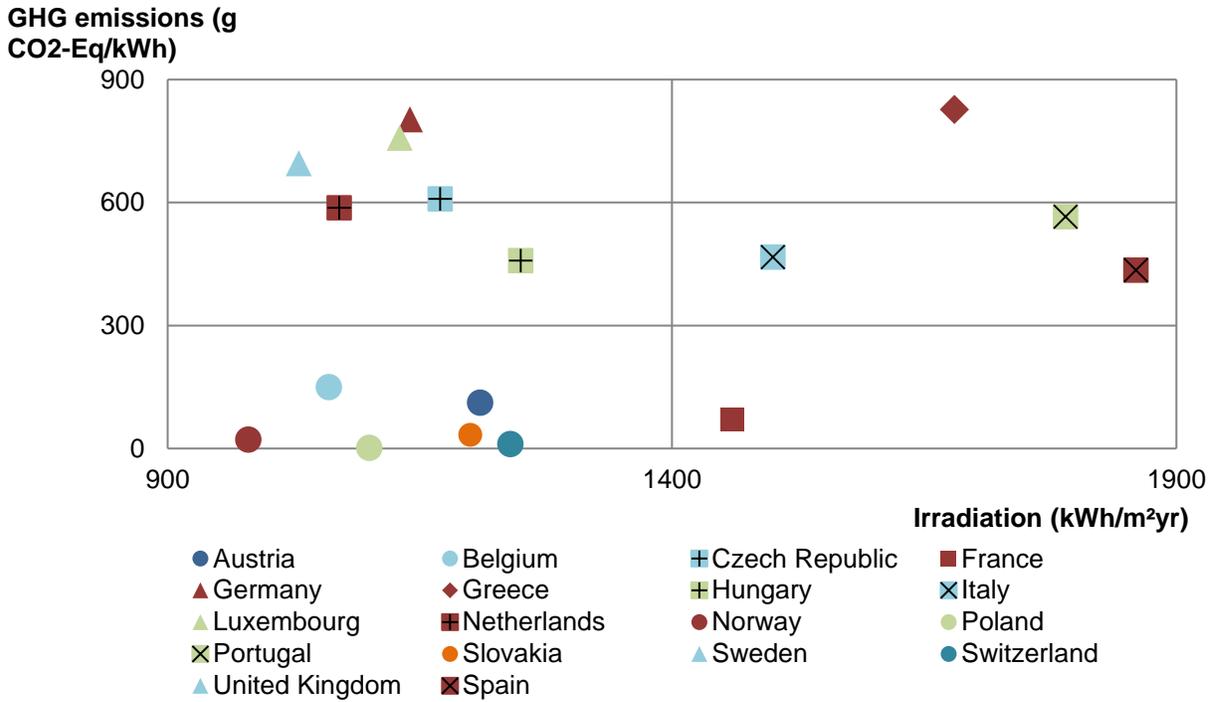


Figure S4-1. Extended comparison between GHG emissions per 1kWh low-voltage electricity and yearly averaged irradiation of 17 countries (out of 23 for which data was available). The data is derived from Table S4-1.

Table S4-4

Absolute impact of production of 10Wh of grid electricity in six countries, indicated in Ecoinvent as “market for electricity, low voltage | conseq. long-term, U“

Impact category	Reference unit	Germany	Spain	France	Greece	NL	Norway
Agricultural land occupation	m <sup>2</sup> *a	-3.58E-03	-1.18E-04	-9.25E-05	-2.57E-04	-1.23E-03	1.40E-05
Climate Change	kg CO <sub>2</sub> eq	8.03E-03	4.35E-03	7.06E-04	8.27E-03	5.87E-03	2.14E-04
Fossil depletion	kg oil eq	1.76E-03	1.08E-03	1.77E-04	2.84E-03	1.66E-03	4.08E-05
Freshwater ecotoxicity	kg 1,4-DB eq	7.55E-04	2.37E-04	1.95E-04	5.85E-04	3.37E-04	1.76E-04
Freshwater eutrophication	kg P eq	2.13E-05	2.32E-06	7.61E-07	2.58E-05	4.16E-06	3.08E-07
Human toxicity	kg 1,4-DB eq	8.32E-03	1.82E-03	9.41E-04	1.58E-02	1.32E-03	5.94E-04
Ionising radiation	kg U235 eq	-1.83E-04	2.31E-03	6.22E-03	2.06E-04	7.08E-04	5.94E-06
Marine ecotoxicity	kg 1,4-DB eq	6.68E-04	2.08E-04	1.70E-04	5.41E-04	2.93E-04	1.52E-04
Marine eutrophication	kg N eq	4.51E-06	1.05E-06	5.89E-07	5.71E-06	1.05E-06	7.48E-08
Metal depletion	kg Fe eq	3.70E-04	2.17E-04	1.79E-04	1.98E-04	2.26E-04	1.36E-04
Natural land transformation	m <sup>2</sup>	4.42E-08	3.67E-07	7.54E-08	3.73E-07	4.69E-07	1.59E-07
Ozone depletion	kg CFC-11 eq	3.62E-11	4.88E-10	9.63E-10	3.93E-10	1.87E-10	1.51E-11
Particulate matter formation	kg PM <sub>10</sub> eq	3.10E-07	1.08E-05	1.60E-06	1.46E-05	1.09E-06	6.07E-07
Photochemical oxidant formation	kg NMVOC	1.34E-05	1.43E-05	2.06E-06	1.31E-05	7.73E-06	5.58E-07
Terrestrial acidification	kg SO <sub>2</sub> eq	9.22E-06	2.97E-05	3.51E-06	3.91E-05	6.15E-06	9.58E-07
Terrestrial ecotoxicity	kg 1,4-DB eq	3.32E-07	2.08E-07	1.29E-07	2.55E-07	1.51E-07	8.06E-08
Urban land occupation	m <sup>2</sup> *a	3.51E-05	3.64E-05	7.35E-06	1.81E-05	3.12E-05	2.58E-06
Water depletion	m <sup>3</sup>	3.87E-02	3.69E-02	4.60E-02	6.22E-02	6.76E-03	1.06E-02

Table S4-5

Absolute impact for production of 10Wp OPV solar charger

Impact category	Reference unit	
Agricultural land occupation	m <sup>2</sup> *a	7.39E-01
Climate Change	kg CO <sub>2</sub> eq	3.89E+00
Fossil depletion	kg oil eq	1.14E+00
Freshwater ecotoxicity	kg 1,4-DB eq	5.84E-02
Freshwater eutrophication	kg P eq	3.67E-04

Human toxicity	kg 1,4-DB eq	1.07E+00
Ionising radiation	kg U235 eq	-1.47E-01
Marine ecotoxicity	kg 1,4-DB eq	4.97E-02
Marine eutrophication	kg N eq	7.19E-04
Metal depletion	kg Fe eq	4.34E-01
Natural land transformation	m2	4.97E-04
Ozone depletion	kg CFC-11 eq	2.52E-07
Particulate matter formation	kg PM10 eq	4.97E-03
Photochemical oxidant formation	kg NMVOC	1.96E-02
Terrestrial acidification	kg SO2 eq	8.10E-03
Terrestrial ecotoxicity	kg 1,4-DB eq	3.59E-03
Urban land occupation	m2*a	2.02E-02
Water depletion	m3	-3.59E+00

## Annex 3: Life cycle assessment of the production of surface-active alkyl polyglycosides from acid-assisted ball-milled wheat straw compared to the conventional production based on corn-starch

### **Abstract**

Production of alkyl polyglycosides from mechanocatalytic depolymerization of wheat straw is a promising route because of the use of an available bio-based feedstock. This study aims to verify the environmental benefit of this process in comparison with a reference process that produces APGs from corn starch-based glucose. Life cycle assessment methodology is used to compare both production routes. The results have shown that the new production route based on wheat straw generate lower environmental impacts compared to the reference process because of the use of wheat straw instead of corn starch-based glucose and the energy recovery from the by-product lignine that meets most of energy demand of the process. The LCA results also show that the production of fatty alcohol dominates the life cycle impacts of APGs. Environmental impacts are sensitive to the source of the fatty alcohol (from palm kernel or coconut oil).

### **Introduction**

Surfactants are chemicals widely used in cosmetic and detergent industries. Sustainability and user-toxicity concerns have prompted shifting demand from synthetic petrochemical to overall safer and bio-based surfactants. In particular, the plant-based surfactants alkyl polyglycosides (APGs) have been particularly promoted (von Rybinski and Hill 1998). Favorable properties such as good wettability, foam production, and cleaning ability have been indicated in application as cleaning agents, as well as dermatological and ocular safety for their use in cosmetics (Pantelic 2014). Due to their perceived environmental safety and compliance to the principles of Green Chemistry, APGs have been considered as green chemicals, and even some APGs have been granted the status of pharmaceutical excipients (Anastas and Eghbali 2010; Guilbot et al. 2013; Pantelic 2014).

Although APGs are bio-based and considered better for the environment in comparison to synthetic surfactants, they are not emission free. Materials and energy inputs needed, and process waste outputs released in manufacture of APGs, result in environmental impacts. In particular, agricultural activities carried out to produce necessary raw materials are particularly impactful. Currently, APGs are synthesized via the Fischer glycosidation, which comprises a reaction between plant-based fatty alcohols (typically sourced from palm or coconut oil) and

carbohydrates in the presence of an acid catalyst. Carbohydrates used are generally refined syrup or powder like glucose coming from corn starch hydrolysis. Cultivation of those cultures are all man-made and require inputs of water, fertilizers, pesticides, harvesting, transportation and other energy and chemical intensive activities.

A newly developed process for manufacturing APGs surfactants using wheat straw could substitute traditional corn-starch-based glucose (Boissou et al. 2015). In acid-assisted ball milling process, APGs are also synthesized via the Fischer glycosidation. However, carbohydrates used in this new process are reactive polysaccharide oligomers coming from acid-catalysed milling of lignocellulosic raw materials. This reactive biomass is directly used in synthesis with fatty alcohols without separation or purification steps. In one hand, this new process allows the valorisation of all polysaccharides available in lignocellulosic raw materials like cellulose and hemicelluloses. This results in the production of alkylpolyglycosides mixture of alkylpolyglucosides and alkylpolypentosides (mainly alkylpolyxylosides). It has been proven that the presence of alkylpolypentosides is responsible for the improved chemical and physical properties and lower toxicity displayed (Marinkovic and Estrine 2010; Martel et al. 2010). On the other hand, non-edible polysaccharides can be used in the production of valuable amphiphilic alkylglycosides and lignin is burnt to provide electricity and heat.

Yet, a current literature on APGs hasn't extensively investigated environmental burdens associated with these APGs. Only few studies attempt to quantify impacts associated with APGs production and use (Guilbot et al. 2013; Lokesh et al. 2017). A recent study analyses the environmental impacts of producing APGs from wheat straw wax extracted with supercritical CO<sub>2</sub> (Lokesh et al. 2017).

In effort to further the knowledge of environmental benefits of APGs and additionally optimise material and energy use in the production of this chemical, the present study investigates environmental impacts of APGs production.

Environmental impact of producing APGs based on wheat straw is therefore investigated, and subsequently compared with conventional corn-starch based APGs. Life cycle assessment (LCA) method was used to account all the inputs of energy and materials and waste emissions in the APGs production and convert them to potential impacts to the environment.

## **Material and methods**

The environmental impacts of the APGs production processes will be based on LCA. LCA is a multicriteria tool to assess the burdens on the environment of a product or a service over its lifetime, i.e., from the production of the raw materials to the end of life management. LCA is suitable to assess the potential benefits of sustainable chemistry (Kralisch et al. 2014) (Kralisch et al. 2014).

The LCA methodology is defined by ISO standards (ISO-14040 2006) and is divided in four steps:

- The goal and scope definition phase: the objectives of the study, the boundaries of the system are clearly presented.
- The inventory analysis phase: the elementary flows (inputs and outputs) of the product system are collected. The inventory entails the quantification of energy, resources, and emissions to air, soil and water.
- The impact assessment phase: based on the inventory, the different flows are converted into environmental impacts.
- The interpretation phase: the results are interpreted and lead to the identification of the environmental hotspots and to recommendations to improve the environmental performance of the product.

### **Goal and scope**

The aim of the study is to investigate environmental performance of alkyl polyglycoside productions using wheat straw as a sugar-base (hydrophilic component) based on a newly developed acid-assisted ball milling process. Impacts associated to wheat straw-based process (“mechanocatalytic process” hereafter) and further compared with the conventional industrial process for manufacturing of APGs using glucose from corn starch (“reference process” hereafter).

The function of the system is the production of APGs, and the corresponding functional unit and reference flow is 1 ton of APGs.

A cradle-to-gate assessment is carried out; production of raw materials and manufacture of APGs are taken into account while use, retail and end-of-life life cycle stages are not included because they are considered similar in both systems.

All data sources and allocation procedures are explained in the LCI section. The LCI is set up in the French context.

Environmental impacts are then characterized using ReCiPe Midpoint (H) life cycle impact assessment method selected as the most up to date method for the European context (Table 1) (Huijbregts et al. 2016). All modelling was done using SimaPro 8.3, LCA software.

Table 1. ReCiPe 2016 midpoint (H) impact categories and list of abbreviations

Impact category	Abbreviations	Unit
Global warming	GW	kg CO <sub>2</sub> eq
Stratospheric ozone depletion	SOD	kg CFC11 eq
Ionizing radiation	IR	kg Co-60 eq
Ozone formation, Human health	OF	kg NO <sub>x</sub> eq
Fine particulate matter formation	FPMF	kg PM2.5 eq
Ozone formation, Terrestrial ecosystems	OF T	kg NO <sub>x</sub> eq
Terrestrial acidification	TA	kg SO <sub>2</sub> eq
Freshwater eutrophication	FEut	kg P eq
Terrestrial ecotoxicity	TE	kg 1,4-DCB eq
Freshwater ecotoxicity	FE	kg 1,4-DCB eq
Marine ecotoxicity	ME	kg 1,4-DCB eq
Human carcinogenic toxicity	HCT	kg 1,4-DCB eq
Human non-carcinogenic toxicity	HNCT	kg 1,4-DCB eq
Land use	LU	m <sup>2</sup> a crop eq
Mineral resource scarcity	MRS	kg Cu eq
Fossil resource scarcity	FRS	kg oil eq
Water consumption	WC	m <sup>3</sup>

## Life Cycle Inventory

### Product system 1 - Mechanocatalytic process

The process flowchart for the mechanocatalytic process is given in Figure 1.

First, wheat straw is grinded in a knife mill and then fed in the ball miller along with sulfuric acid to release and convert cellulose and hemicelluloses to short chain oligosaccharides. Then, glycosylation process is carried in the reaction between the mechanocatalytically depolymerized

cellulose and fatty alcohols either from palm kernel or coconut oil. The resulting products are lignin, APGs (as a mixture of alkylpolyglucosides and alkylpolypentosides) and sodium sulfate (Boissou et al. 2015).

Grinding, ball milling and glycosylation are modelled as foreground processes with data collected in the literature for the energy use and at the lab-scale for the quantities (Boissou et al. 2015). Data are reported in Figure 1.

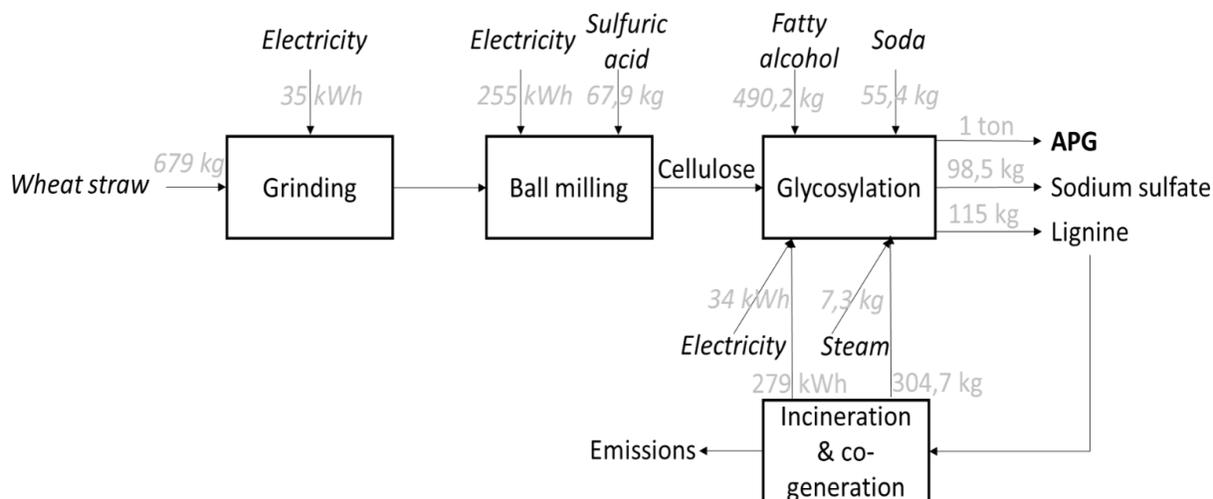


Figure 1. Process flow chart of the mechanocatalytic process

Energy use for grinding wheat straw having size of 0.8mm has been reported to be 51.6 kWh/t of wheat straw (Tumuluru et al. 2014). resulting in 35 kWh/t of APG. As for the ball milling process, Kaufman Rechulski et al. (2015) reported an energy use of 376 kWh/t (Kaufman Rechulski et al. 2015), resulting in 255 kWh/t of APG.

Energy use for glycosylation was taken from Guilbot et al. (2013) that report 313 kWh/t of APG for electricity (pumps, stirring motors) and 104 kg of steam/t of APG for the reaction. We added an extra consumption of 208 kg of steam for recovering fatty alcohol in excess from the system by distillation. The total energy demand of the glycosylation process is 313 kWh of electricity and 312 kg of steam. For this process, we additionally assumed the incineration of the co-produced lignin with energy recovery in a cogeneration unit and energy reinjected back in the system to meet most of the energy demand of the glycosylation. The cogeneration unit allows the recovery of 115kg of lignin per ton of APGs produced. This is equivalent to 2875MJ/ton of APGs considering a lower heating value of the lignin equal to 25kJ/kg (Sheng and Azevedo 2005). In the cogeneration unit, 35% of the total energy can be recovered in electricity, i.e., 1006.25MJ or 279 kWh. The remaining energy can be valorised to produce steam. We assumed that half of the remaining energy is converted in steam, i.e., 934.4 MJ. The production of 1 kg of steam for

chemical industry needs 3.067 MJ of energy according to ecoinvent data. Therefore 304.7 kg of steam can be produced per ton of APGs. At last the glycosylation process only requires additional energy input of 34kWh of electricity and 7.3 kg of steam (Figure 1).

For the remaining byproducts (APGs and sodium sulfate) an allocation of the burdens had to be made. For the baseline scenario, an economic allocation is done to share the impacts between the APGs and the sodium sulfate. The economic allocation calculation is given by the following formula:

$$A_{APG} = \frac{m_{APG} \cdot P_{APG}}{m_{APG} \cdot P_{APG} + m_{sodium\_sulfate} \cdot P_{sodium\_sulfate}}$$

Where A is the allocation factor, m the mass (t) and P the price (€/t).

The prices were given by an expert source: 2 500 and 300 €/t for APGs and sodium sulfate respectively. For the production of 1 ton of APGs, 98.5 kg of sodium sulfate are obtained. Thus, APGs accounts for 99% of the burdens of the overall process.

Figure 1 gives an overview of material and energy inputs for the mechanocatalytic process.

The LCI concerning the background production of energy and reactants (in italic) were taken from Ecoinvent v3.3 database (Wernet et al. 2016). Table S1 in Supplementary information gives the background processes names from ecoinvent used for the ball milling process.

The study has been made in a French context: all the data used is considered as the most accurate for the French situation. For example, the straw coming from wheat production is a Swiss process. Both countries have similar agricultural systems.

### **Product system 2 - reference process**

The process flowchart and inventory data for the reference process is given in Figure 2 taken from (Hirsinger 1997). The APGs (mainly alkylpolyglucosides) are obtained via Fischer Glycosylation process in reaction between powder glucose, fatty alcohols and sulfuric acid. Sodium sulfate is also generated after neutralization step at a very low level (less than 1wt%/APG). It is included to the final product.

The energy for the glycosylation reported in Hirsinger et al. (1997) is 3250 kWh which is more than 6 times higher than for the glycosylation step of the mechanocatalytic process, probably because the process is outdated. As a matter of consistency, we assumed a similar energy demand in both processes, i.e., 313 kWh of electricity and 312 kg of steam.

Background data are taken from ecoinvent 3.3. We considered corn starch background process instead of corn starch-based glucose. This is because ecoinvent does not provide data for the latter process. The hydrolysis of corn starch to glucose has a small share of impact in the life cycle of this feedstock, and according to Hirsinger et al. (1997) the reference process can also use corn starch. Table S2 in Supplementary information gives the the background processes names used for reference process.

## Results

### Process contribution analysis of the mechanocatalytic process

In this section, the life cycle impact assessment results of the acid-assisted ball milling process are presented in Figure 3 (a, b), and absolute values are provided in Supplementary Information (Table S4).

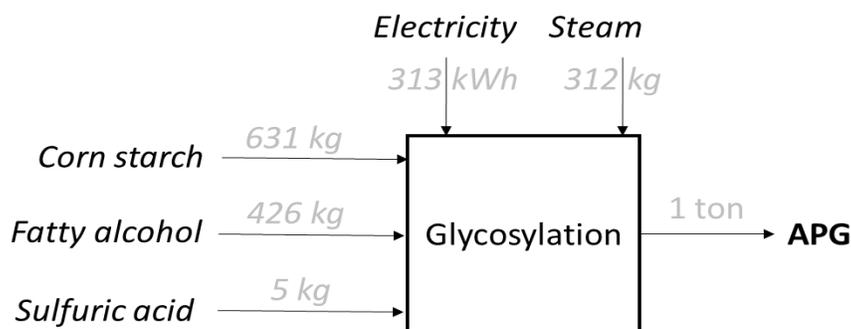


Figure 2. Process flowchart of reference process - APGs production from corn-starch

The results show that, in average, 90 % of the impacts come from the production of the fatty alcohols. The second main raw material, i.e., wheat straw accounts for a small share of the impacts. The mechanocatalytic process only accounts for few impacts. One exception can be noticed regarding the ionizing radiation indicator. In this case, the electricity used during the process has the highest share of impacts because of the French electricity mix mainly composed of nuclear energy.

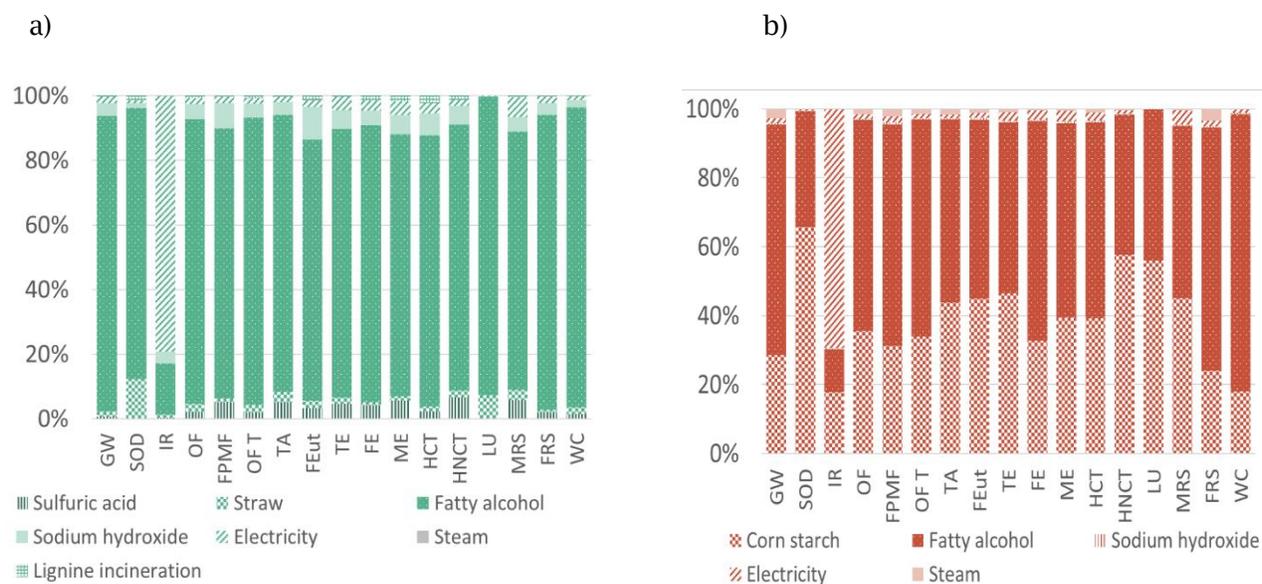


Figure 3 (a, b). a) Contribution analysis of APGs production with mechanocatalytic process b) Contribution analysis of reference process for APGs production. Abbreviations definitions are available in Table 1 (Huijbregts et al. 2016).

### Process contribution analysis of reference process

The process contribution of the reference process on the environmental impacts is given in Figure 4.

It highlights that, similarly to the mechanocatalytic process, the main burdens come from the production of the fatty alcohols. However, the feedstock corn starch has also important contribution in most of impact categories. The specific electricity impacts from nuclear energy can be observed in the ionizing radiation indicator for the same reasons exposed before.

### Comparison between mechanocatalytic and reference process

FiComparison between the mechanocatalytic and the reference processes is provided in Figure 5 (a, b). Absolute values are provided in Supplementary Information (Table S4)

Mechanocatalytic process results in lower impacts across all categories. This is mainly due to the use of wheat straw in the mechanocatalytic process that generate a small share of impacts compared to corn starch in the reference process. Also, the recovered energy from lignin in the mechanocatalytic process lowers the energy demand compared to the reference process. Even if the quantity of fatty alcohols to produce 1ton APGs with the mechanocatalytic process is higher compared to the reference process (490.2kg in comparison to 426kg), influence of corn starch and electricity related impacts affect overall result in favour of the mechanocatalytic process.

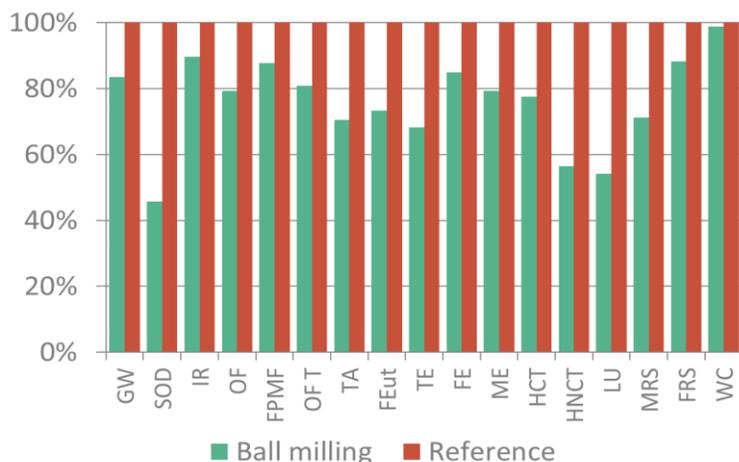


Figure 5. Environmental impact comparison between mechanocatalytic and reference process.

## Discussions

### Influence of the allocation procedure

The share of the burdens between the sodium sulfate and the APGs for the mechanocatalytic process was made using an economic allocation. A mass allocation is tested with the following formula:

$$A_{APG} = \frac{m_{APG}}{m_{APG} + m_{sodium\_sulfate}}$$

The quantity of APGs and sodium sulfate are 1 t and 98 kg respectively thus allocating 91 % of the environmental impacts to the surfactants. The environmental burdens of the surfactant are higher in the case of the economic allocation because prices lead to a higher share (99 %) that a

mass allocation (91%). However, the choice of the allocation procedure has a low influence in the final results.

### Influence of the source of fatty alcohols

The selected LCI background data for fatty alcohols (“Fatty alcohol {GLO}market for”) is a global market dataset comprising three means of production: palm kernel oil, coconut oil and petrochemicals. As fatty alcohol contributes to a large share of impact in both processes, we made a sensitivity analysis to analyse the influence of the alcohol production route on the impacts of APGs production (Figure 6).

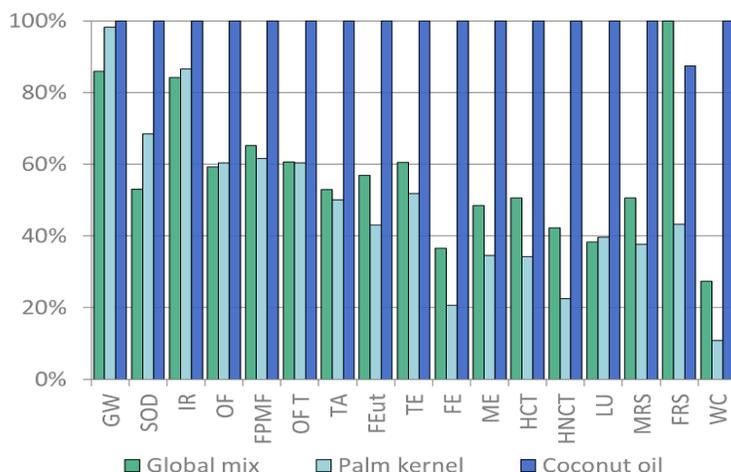


Figure 6. Sensitivity analysis from 3 fatty alcohol production sources for the mechanocatalytic process

The comparison between the 3 datasets shows that the use of fatty alcohols from coconut oil generates the largest important environmental burdens for most of impact categories. In general, the palm kernel source presents fewer impacts than the global market dataset.

The difference between palm kernel oil and coconut oil production is explained by the fact that the production of coconut is a more pollutant activity. For example, in the Terrestrial Acidification indicator, the quantity of ammonia is 4 times higher in the case of the coconut production.

### Influence of the electricity mix

This study has been realised in a French context so we used the French electricity mix. It appeared interesting to show the influence of the spatial localisation. In other words, we did the modelling using German and the European electricity mix. The results are shown in Figure 7.

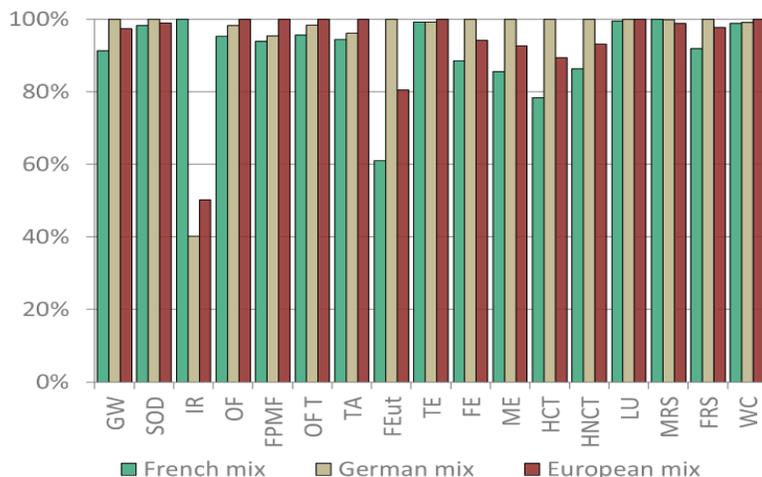


Figure 7. Sensitivity analysis from 3 electricity mixes for the mechanocatalytic process

The use of French electricity induces, in most of the indicators, less impact. The French electricity mix scenario generates higher ionizing radiation impacts because of the emissions of radionuclides (such as Radon-222) during the extraction of the nuclear fuel and the operation of the nuclear plants.

However, the difference is, in average, less than 10 %. It is explained by the fact that most of the impacts come from the production of fatty alcohols independently from the electricity mix.

### Comparison with other studies

The concern about the environmental impact of the surfactant has arisen during the last few years. Recently, two studies have been published dealing with the environmental impacts of alkyl polyglucosides (Guilbot et al. 2013; Lokesh et al. 2017). In the first study, the authors conducted the LCA of a cosmetic cream composed of APGs. The APGs are a combination of cetearyl alcohol (80%) and cetearyl glucoside. The authors also found that the most contributing phase is the production of the fatty alcohols in most of their selected impact categories considered: ozone depletion, global warming, mineral resources, petrochemical resources, eco-toxicity and acidification/eutrophication. It emphasizes the fact that the source of fatty alcohol is of great importance. Moreover, the surfactants are based on starch from wheat grain whereas the mechanocatalytic process used wheat straw, a low-value and non-edible resource. Finally, climate

change impacts of wheat straw based and cetearyl glucoside/alcohol-based products can be compared with keeping in mind that they rely on different assumptions: 1.8 and 12.4 kg of emitted CO<sub>2</sub> eq respectively. It shows again the environmental benefits of the mechanocatalytic process compared to a conventional one.

The second study (Lokesh et al. 2017), assesses the environmental performance of APGs sourced from wheat straw and transformed with supercritical CO<sub>2</sub>. The impacts of the production of APGs are quantified using 5 indicators: direct GHG emissions, land use change & emissions, fossil derived energy footprint, water consumption and a waste factor. These indicators, at the exception of the GHG emissions, can be described as inventory indicators rather than environmental impact indicators. A comparison between the mechanocatalytic process and the supercritical CO<sub>2</sub> process shows that the emissions are in the same order of magnitude: 1.8 and 1.6 kg CO<sub>2</sub> eq respectively. However, the comparison should also be done with precaution because both studies rely on different assumptions and background processes, especially for the agriculture processes.

## **Conclusions**

This paper proposes to assess the environmental impacts of a promising new route for producing APGs. The LCA showed that the use of wheat straw under current assumptions of material input lowers the environmental impacts of APGs compared to a reference scenario using corn starch. The mechanocatalytic process seems to be a more desirable way to produce APGs. The production chain has been thought to reduce the environmental impacts of the production of surfactants. First, the raw material i.e. wheat straw is a residue from wheat production with limited value either in terms of economic value or environmental burdens, and it generates less impacts than glucose in conventional starch corn derived processes. Second, the recovery of lignin into electricity and steam meets 34% of the electricity demand and 98% of the heat demand of the whole process. This study has also pointed out that the environmental burdens of the surfactants come from the production of fatty alcohols. This tendency can be observed in all the indicators except for the ozone depletion and the ionizing radiation where the production of electricity in the nuclear plants is responsible for the impacts. Different sensitivity analyses were carried out on three parameters: allocation procedure, fatty alcohols sources and electricity mix. The allocation procedure does not change the outcomes so either allocation can be chosen. The electricity mix has a logical influence on the indicators that are driven by the energy inputs. We observed that the mechanocatalytic process had less impacts with French electricity compared to German or European electricity except for ionizing radiation. However, the energy required for the grinding and the glycosylation processes have a small contribution in the environmental impacts. Thus, the choice of the electricity mix has a limited influence on the results. The source

for the fatty alcohols production is the main source of variability as fatty alcohols are the main impact contributors.

### Supplementary Information:

Table S1. Foreground data and background data sources for the ball milling process

Flow/Process	Quantity	Ecoinvent process (background data source)
Straw	679 kg	Straw {CH}  wheat production, Swiss integrated production, intensive   Alloc Def
Sulfuric acid	67,9 kg	Sulfuric acid {GLO}  market for   Alloc Def
Fatty alcohols	490,2 kg	Fatty alcohol {GLO}  market for   Alloc Def
Steam	7,3 kg	Steam, in chemical industry {RER}  production   Alloc Def
Soda	55,4 kg	Sodium hydroxide, without water, in 50% solution state {GLO}  market for   Alloc Def
Electricity	35 kWh (grinding) 255 kWh (ball milling) 34 kWh (glycosylation)	Electricity, medium voltage {FR}  market for   Alloc Def
Lignin incineration	115 kg	Biowaste {GLO}  treatment of biowaste, municipal incineration   Alloc Def

Table S1. Foreground data and background data sources for the reference process

Flow/Process	Quantity	Ecoinvent or GaBi process (background data source)
Corn starch	631 kg	Maize starch {GLO}  market for   Alloc Def
Fatty alcohol	426 kg	Fatty alcohol {GLO}  market for   Alloc Def
Sulfuric acid	5 kg	Sulfuric acid {GLO}  market for   Alloc Def
Electricity	313 kWh	Electricity, medium voltage {FR}  market for   Alloc Def
Steam	312 kg	Steam, in chemical industry {RER}  production   Alloc Def

Table S3. Additional processes used for the sensitivity analysis

Flow/Process	Ecoinvent or GaBi process (background data source)
Fatty alcohol from coconut oil	Fatty alcohol {RER}  production, from coconut oil   Alloc Def
Fatty alcohol from palm kernel	Fatty alcohol {RER}  production, from palm kernel oil   Alloc Def
Electricity german mix	Electricity, medium voltage {DE}  market for   Alloc Def
Electricity European mix	Electricity, medium voltage {Europe without Switzerland}  market group for   Alloc Def

Table S4. Absolute impact results for ball milling and reference processes (ReCiPe2016 Midpoint H) - 1 ton of APG

<b>Impact category</b>	<b>Unit</b>	<b>Ball milling process</b>	<b>Reference process</b>
Global warming	kg CO2 eq	1870.856	2240.740
Stratospheric ozone depletion	kg CFC11 eq	0.005	0.010
Ionizing radiation	kBq Co-60 eq	236.455	263.685
Ozone formation, Human health	kg NOx eq	3.803	4.800
Fine particulate matter formation	kg PM2.5 eq	2.411	2.748
Ozone formation, Terrestrial ecosystems	kg NOx eq	4.138	5.123
Terrestrial acidification	kg SO2 eq	8.315	11.786
Freshwater eutrophication	kg P eq	0.406	0.554
Terrestrial ecotoxicity	kg 1,4-DCB e	0.908	1.333
Freshwater ecotoxicity	kg 1,4-DCB e	52.735	62.124
Marine ecotoxicity	kg 1,4-DBC e	54.843	69.254
Human carcinogenic toxicity	kg 1,4-DBC e	42.479	54.840
Human non-carcinogenic toxicity	kg 1,4-DBC e	41108.948	72891.338
Land use	m2a crop eq	1053.839	1945.836
Mineral resource scarcity	kg Cu eq	4.563	6.407
Fossil resource scarcity	kg oil eq	493.554	559.275
Water consumption	m3	96.307	97.476